

Physics 673: Quantum Optics and Nonlinear Optics

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University of Calgary, Winter 2007

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Chapter 1

Maxwell's Equations and Gauge Fields

1.1 Maxwell's Equations

describe all phenomena related to (classical) electrical and magnetical fields. In *free space* they are given by:

$$\begin{aligned}\operatorname{div} \mathbf{E} &= \frac{\varrho}{\varepsilon_0} & \operatorname{div} \mathbf{B} &= 0 \\ \operatorname{curl} \mathbf{E} &= -\dot{\mathbf{B}} & \operatorname{curl} \mathbf{B} &= \mu_0(\mathbf{j} + \varepsilon_0 \dot{\mathbf{E}})\end{aligned}\tag{1.1}$$

where \mathbf{E} denotes the electric and \mathbf{B} the magnetic field; $\varrho(\mathbf{x}, t)$ is the charge density and $\mathbf{j}(\mathbf{x})$ current density. The divergence and the rotation of a function $\mathbf{R}(\mathbf{x})$ is defined by

$$\begin{aligned}\operatorname{div} \mathbf{R}(\mathbf{x}) &= \sum_{i=1}^3 \frac{\partial}{\partial x_i} R_i(\mathbf{x}) = \partial_i R_i(\mathbf{x}) \\ (\operatorname{curl} \mathbf{R}(\mathbf{x}))_i &= \sum_{j,k=1}^3 \epsilon_{ijk} \frac{\partial}{\partial x_j} R_k = \epsilon_{ijk} \partial_j R_k(\mathbf{x})\end{aligned}$$

The notation on the very right-hand side employs Einstein's summation convention: summation (here from 1 to 3) is implied for each index that appears twice ϵ_{ijk} is the antisymmetric Levi-Civita-Symbol, defined by

$$\epsilon_{ijk} = \begin{cases} 1 & (ijk) = (123), (231), (312) \\ -1 & (ijk) = (132), (321), (213) \\ 0 & \text{otherwise} \end{cases}$$

It follows that $\epsilon_{ijk} = \epsilon_{jki}$ and $\epsilon_{ijk} = -\epsilon_{jik}$.

For consistency the *continuity equation* must be valid:

$$\dot{\varrho} + \operatorname{div} \mathbf{j} = 0 \quad (1.2)$$

Proof:

$$\begin{aligned} \dot{\varrho} &= \varepsilon_0 \operatorname{div} \dot{\mathbf{E}} \\ &= \varepsilon_0 \operatorname{div} \left(\frac{1}{\mu_0 \varepsilon_0} (\operatorname{curl} \mathbf{B} - \mu_0 \mathbf{j}) \right) \\ &= \frac{1}{\mu_0} \operatorname{div} \operatorname{curl} \mathbf{B} - \operatorname{div} \mathbf{j} \\ \operatorname{div} \operatorname{curl} \mathbf{B} &= \partial_i \epsilon_{ijk} \partial_j B_k = \underbrace{\epsilon_{ijk}}_{\text{antisym.}} \underbrace{\partial_i \partial_j}_{\text{sym.}} B_k = 0 \end{aligned} \quad (1.3)$$

The physical meaning of the continuity equation is charge conservation:

$$\begin{aligned} Q(t) &= \int_V \varrho(\mathbf{x}, t) d^3x \\ \implies \dot{Q} &= \int_V \dot{\varrho} d^3x = - \int_V \operatorname{div} \mathbf{j} d^3x \end{aligned}$$

Using Gauss' theorem,

$$\int_V \operatorname{div} \mathbf{R} d^3x = \oint_{\partial V} \mathbf{R} \cdot d\mathbf{s} \quad (1.4)$$

one can see that the change of the total charge inside a volume V is equal to the total current which flows out of it:

$$\dot{Q} = - \oint_{\partial V} \mathbf{j} \cdot d\mathbf{s} \quad (1.5)$$

Here ∂V denotes the surface (= boundary) of V .

We now will derive differential equations of second order from Maxwell's equations:

$$\begin{aligned} (\operatorname{curl} \operatorname{curl} \mathbf{E})_i &= \epsilon_{ijk} \partial_j (\operatorname{curl} \mathbf{E})_k \\ &= \epsilon_{ijk} \partial_j \epsilon_{klm} \partial_l E_m \end{aligned}$$

Exploiting $\epsilon_{ijk} \epsilon_{klm} = \delta_{il} \delta_{jm} - \delta_{im} \delta_{jl}$, one finds

$$(\operatorname{curl} \operatorname{curl} \mathbf{E})_i = \delta_{il} \delta_{jm} \partial_j \partial_l E_m - \delta_{im} \delta_{jl} \partial_j \partial_l E_m$$

$$\begin{aligned}
\Rightarrow (\text{curl curl } \mathbf{E})_i &= \partial_i \partial_m E_m - \partial_l \partial_l E_i \\
&= \nabla_i \text{div } \mathbf{E} - \Delta E_i \\
\text{curl curl } \mathbf{E} &= \nabla \text{div } \mathbf{E} - \Delta \mathbf{E}
\end{aligned}$$

$$\begin{aligned}
\Rightarrow \nabla \text{div } \mathbf{E} - \Delta \mathbf{E} &= -\text{curl } \dot{\mathbf{B}} = -\partial_t \mu_0 (\mathbf{j} + \varepsilon_0 \dot{\mathbf{E}}) \\
\Rightarrow \frac{1}{\varepsilon_0} \nabla \varrho - \Delta \mathbf{E} &= -\mu_0 \partial_t \mathbf{j} - \varepsilon_0 \mu_0 \ddot{\mathbf{E}}
\end{aligned}$$

Using

$$\varepsilon_0 \mu_0 = \frac{1}{c^2}$$

it follows that

$$\boxed{\left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right) \mathbf{E} = -\mu_0 \frac{\partial}{\partial t} \mathbf{j} - \frac{1}{\varepsilon_0} \nabla \varrho} \quad (1.6)$$

The d'Alembert operator $\left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right)$ is often represented by \square . Similarly one can show that

$$\begin{aligned}
\ddot{\mathbf{B}} &= -\text{curl } \dot{\mathbf{E}} = -\text{curl } \frac{1}{\varepsilon_0 \mu_0} (\text{curl } \mathbf{B} - \mu_0 \mathbf{j}) \\
\Rightarrow \frac{1}{c^2} \ddot{\mathbf{B}} &= -(\nabla \underbrace{\text{div } \mathbf{B}}_{=0} - \Delta \mathbf{B}) + \mu_0 \text{curl } \mathbf{j} \\
\Rightarrow \boxed{\left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right) \mathbf{B} &= \mu_0 \text{curl } \mathbf{j}} \quad (1.7)
\end{aligned}$$

These equations are *wave equations*! One can distinguish between several special cases:

1. $\mathbf{j}, \varrho = 0 \Rightarrow$ free propagation of waves, the differential equations are linear and homogeneous.
2. \mathbf{j}, ϱ are given functions \Rightarrow inhomogeneous differential equations describing the electromagnetic fields caused by currents and charges.
3. \mathbf{j}, ϱ depend (approximately) linearly on \mathbf{E}, \mathbf{B} .
 \Rightarrow linear dielectric media (polarisable media), described by linear differential equations.
4. \mathbf{j}, ϱ depend nonlinearly on \mathbf{E}, \mathbf{B} .
 \Rightarrow nonlinear differential equations, generally hard to solve (Nonlinear Optics).

1.2 Scalar Potential and Vector Potential

Maxwell's equation (1.1) states that $\text{div } \mathbf{B} = 0$ ($= \partial_i B_i$). According to Eq. (1.3) we generally have $\text{div } \text{curl } \mathbf{A} = 0$. We therefore can write the magnetic field in the form ¹

$$\boxed{\mathbf{B} = \text{curl } \mathbf{A}} \quad \mathbf{A} = \text{Vector potential}$$

\mathbf{B} does not possess any sources (in some speculative theories such sources are considered and represent magnetic Monopoles).

Inserting this into (1.1) we find

$$\Rightarrow \text{curl } \mathbf{E} = -\dot{\mathbf{B}} = -\text{curl } \dot{\mathbf{A}}$$

$$\Rightarrow \text{curl } (\mathbf{E} + \dot{\mathbf{A}}) = 0$$

we have generally $\text{curl } \text{grad } \phi = \epsilon_{ijk} \partial_j \partial_k \phi = 0$

$$\Rightarrow \mathbf{E} + \dot{\mathbf{A}} = -\nabla \phi \quad \text{in a simply connected space}$$

$$\Rightarrow \boxed{\mathbf{E} = -\nabla \phi - \dot{\mathbf{A}}} \quad \phi = \text{scalar Potential}$$

The potentials \mathbf{A} and ϕ are not measurable, in contrast to \mathbf{E} and \mathbf{B} . The equations of motion for the potentials are given by

$$\text{div } \mathbf{E} = \frac{\rho}{\epsilon_0} = -\text{div } \text{grad } \phi - \text{div } \dot{\mathbf{A}}$$

$$\text{curl } \mathbf{B} = \text{curl } \text{curl } \mathbf{A} = \mu_0 (\mathbf{j} + \epsilon_0 [-\nabla \dot{\phi} - \ddot{\mathbf{A}}])$$

$$\Rightarrow \boxed{\begin{aligned} \Delta \phi &= -\frac{\rho}{\epsilon_0} - \text{div } \dot{\mathbf{A}} \\ \left(\frac{1}{c^2} \ddot{\mathbf{A}} - \Delta \mathbf{A} \right) + \nabla \text{div } \mathbf{A} &= \mu_0 \mathbf{j} - \frac{1}{c^2} \nabla \dot{\phi} \end{aligned}} \quad (1.8)$$

If $\text{div } \mathbf{A} = 0$ would hold, ϕ would obey Poisson's equation and the evolution of \mathbf{A} would be governed by a wave equation. In this case (!) ϕ represents the usual Coulomb potential. We will see below how to achieve this.

1.3 Gauge transformations

$\mathbf{B} = \text{curl } \mathbf{A}$ does not fix \mathbf{A} completely. Because $\text{curl } \text{grad } \chi = 0$ holds for any χ , the vector potential

$$\mathbf{A}' = \mathbf{A} + \nabla \chi$$

¹We disregard subtle topological effects which appear in multiply connected spaces.

does lead to the same magnetic field. Likewise, ϕ and

$$\phi' = \phi - \partial_t \chi$$

create the same electric field since

$$\begin{aligned} \mathbf{E}' &= -\nabla \phi' - \dot{\mathbf{A}}' = -\nabla \phi + \nabla \dot{\chi} - \dot{\mathbf{A}} - \nabla \dot{\chi} \\ &= -\nabla \phi - \dot{\mathbf{A}} = \mathbf{E} \end{aligned}$$

These transformations are called Gauge transformations. The physical fields \mathbf{E} , \mathbf{B} are not changed by a gauge transformation. Gauge transformations can be used to impose certain (convenient) constraints, called gauges, on the potentials. The most common gauges are

| | |
|--|---------------|
| $\text{div} \mathbf{A} = 0$ | Coulomb gauge |
| $\text{div} \mathbf{A} + \frac{1}{c^2} \frac{\partial}{\partial t} \phi = 0$ | Lorentz gauge |

(1.9)

Both are independent from each other and generally cannot be fulfilled simultaneously. Coulomb gauge leads to the Coulomb potential but is not covariant under Lorentz transformations. Lorentz gauge is covariant and therefore very important for relativistic situations.

Remarks on the derivation of Coulomb gauge:

Let \mathbf{A}' , ϕ' be some potentials. We need to find a function χ so that $\text{div} \mathbf{A} = 0$ after a gauge transformation:

$$\mathbf{A} = \mathbf{A}' - \nabla \chi \implies \text{div} \mathbf{A} = \text{div} \mathbf{A}' - \Delta \chi \stackrel{!}{=} 0$$

This is a differential equation for χ ,

$$\Delta \chi = \text{div} \mathbf{A}' \quad \text{Poisson equation for } \chi, \quad (1.10)$$

with the solution

$$\chi(\mathbf{x}, t) = -\frac{1}{4\pi} \int \frac{\text{div} \mathbf{A}'(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} d^3 x'$$

Proof:

$$\Delta_x \chi = -\frac{1}{4\pi} \int \text{div} \mathbf{A}'(\mathbf{x}') \Delta_x \frac{1}{|\mathbf{x} - \mathbf{x}'|} d^3 x'$$

We now use the very important theorem

| |
|--|
| $\Delta_x \frac{1}{ \mathbf{x} - \mathbf{x}' } = -4\pi \delta(\mathbf{x} - \mathbf{x}')$ |
|--|

$$\begin{aligned} \Rightarrow \quad \Delta_x \chi &= -\frac{1}{4\pi} \int \operatorname{div} \mathbf{A}'(\mathbf{x}') (-4\pi) \delta(\mathbf{x} - \mathbf{x}') \\ &= \operatorname{div} \mathbf{A}'(\mathbf{x}) \end{aligned} \quad \text{qed.}$$

Inserting the Coulomb gauge Eq. (1.9) into Eq. (1.8) leads to the field equations for the potentials in Coulomb gauge,

$$\boxed{\begin{aligned} \left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right) \mathbf{A} &= \mu_0 \mathbf{j} - \frac{1}{c^2} \nabla \dot{\phi} \\ \Delta \phi &= -\frac{\rho}{\epsilon_0} \end{aligned}} \quad (1.11)$$

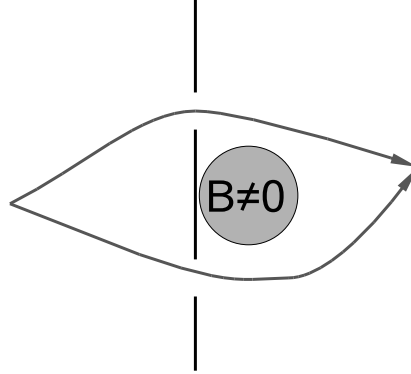
Interpretation: $\phi \hat{=}$ Coulomb potential

$\mathbf{A} \hat{=}$ electromagnetic waves

Attention: the physical interpretation of the scalar and vector potential depends on the gauge condition!

1.3.1 Aharonov-Bohm effect

The Aharonov-Bohm effect [42] is one of the most intriguing effects in quantum mechanics and demonstrates how subtle nature can be. Consider an electron interference experiment where the electrons are moving around a solenoid (see figure)



Inside the solenoid the magnetic field is nonzero, but the electrons are confined to move in the region where the magnetic field vanishes. We therefore would expect that the solenoid does not generate a phase shift in the electron's interference pattern. However, this is not the case. A vector potential that corresponds to a vanishing magnetic field away from the origin is given by

$$\mathbf{A}_{AB} = \frac{\Phi}{r} \mathbf{e}_\phi \quad , \quad \nabla \times \mathbf{A}_{AB} = 0 \text{ for } r \neq 0 \quad (1.12)$$

with $r = \sqrt{x^2 + y^2}$ and $\mathbf{e}_\phi = (-\sin \phi, \cos \phi, 0)$ where ϕ is the angle of cylindrical coordinates in the x-y plane.

The phase shift induced by a vector potential can be calculated using the WKB approximation (see, e.g., chapter 9 of Ref. [43]) and is given by $\Delta\varphi = q/\hbar \oint \mathbf{A} \cdot d\mathbf{x}$. If we integrate along a circular trajectory we have $d\mathbf{x} = r\mathbf{e}_\phi d\phi$ and hence

$$\begin{aligned}\Delta\varphi &= \frac{q}{\hbar} \int_0^{2\pi} r d\phi \mathbf{A}_{AB} \cdot \mathbf{e}_\phi \\ &= \frac{2\pi q\Phi}{\hbar}.\end{aligned}\tag{1.13}$$

Now it seems that the solenoid induces a phase shift even though the electrons always move in a field-free region. To make matters worse we can apparently (!) change this result by applying a gauge transformation: the new vector potential

$$\mathbf{A}' = \mathbf{A} + \nabla\chi \quad \text{with } \chi = -\Phi\phi\tag{1.14}$$

seems to be zero everywhere, so the phase shift calculated with \mathbf{A}' should be zero, too.

The resolution of this paradox is subtle. First of all, the phase shift (1.13) has been experimentally verified shortly after its prediction. But why is the phase shift predicted with the aid of \mathbf{A}' wrong? The answer is that \mathbf{A}' cannot be set to zero everywhere because the gauge function χ is ill defined at the point $\phi = 0 = 2\pi$. In a proper treatment, which is mathematically advanced, a change of gauge that is defined everywhere will not change the AB phase shift. More on this can be found in Refs. [44, 45].

To conclude this section I also want to mention that the AB effect is an example of a topological effect. The phase shift does obviously not depend on the radius of the circular trajectory chosen. In fact, it can be shown that $\Delta\varphi$ does not depend on the shape of the trajectory at all as long as it is enclosing the solenoid. Effects that do not depend on smooth changes of the shape of an object but depend on its overall structure are called topological. Typically topological effects appear when the space in which particles can move are multiply connected. In the AB effect an electron can take two principal ways around the solenoid (left and right) so that the space in which it can move is two-fold connected. This is the mathematical reason behind the AB effect.

1.4 Multipole Expansion

Multipoles are a *very* important tool in Electrodynamics. They are, for instance, used for the derivation of selection rules of quantum mechanical atoms and molecules.

Basically the multipole expansion is an expansion in terms of angular momentum eigenfunctions very similarly to quantum mechanics.

Scalar Multipoles

Example: scalar potential in Lorentz gauge.

$$\square \phi(\mathbf{x}, t) = \frac{\varrho(\mathbf{x}, t)}{\varepsilon_0}$$

Fourier transformation in time leads to

$$\begin{aligned} \tilde{\phi}_\omega(\mathbf{x}) &= \int e^{-i\omega t} \phi(\mathbf{x}, t) dt \\ \implies (\Delta + k^2) \tilde{\phi}_\omega(\mathbf{x}) &= -\frac{\tilde{\varrho}(\mathbf{x})}{\varepsilon_0} \quad , \quad k = \frac{\omega}{c} \end{aligned}$$

This is the inhomogeneous Helmholtz equation. Green's function for Helmholtz's equation fullfills

$$(\Delta + k^2)G(\mathbf{x}, \mathbf{x}') = -\delta(\mathbf{x} - \mathbf{x}')$$

For the boundary condition $G \rightarrow 0$ for $|\mathbf{x} - \mathbf{x}'| \rightarrow \infty$ it is given by

$$\begin{aligned} G(\mathbf{x} - \mathbf{x}') &= \frac{1}{4\pi} \frac{e^{ik|\mathbf{x} - \mathbf{x}'|}}{|\mathbf{x} - \mathbf{x}'|} \\ &= ik \sum_{l=0}^{\infty} j_l(kr_{<}) h_l^{(1)}(kr_{>}) \sum_{m=-l}^m Y_{lm}^*(\vartheta', \phi') \underbrace{Y_{lm}(\vartheta, \phi)}_{\text{basis function}} \end{aligned}$$

where

$$j_l = (-x)^l \left(\frac{1}{x} \partial_x \right)^l \frac{\sin x}{x} \quad \text{spherical Bessel functions} \quad (1.15)$$

$$h_l^{(1)} = j_l - (-x)^l \left(\frac{1}{x} \partial_x \right)^l \frac{\cos x}{x} \quad \text{spherical Hankel functions} \quad (1.16)$$

The solution is given by

$$\begin{aligned} \tilde{\phi}_\omega(\mathbf{x}) &= \int G(\mathbf{x}, \mathbf{x}') \frac{\tilde{\varrho}(\mathbf{x}')}{\varepsilon_0} d\Omega' \\ \implies \tilde{\phi}_\omega(\mathbf{x}) &= \frac{ik}{\varepsilon_0} \int \sum_{l=0}^{\infty} \sum_{m=-l}^l j_l(kr_{<}) h_l^{(1)}(kr_{>}) Y_{lm}(\vartheta, \varphi) dr' \\ &\quad \cdot \int Y_{lm}^*(\vartheta', \varphi') \tilde{\varrho}_\omega(r', \vartheta', \varphi') d\Omega' \end{aligned}$$

Particularly important for practical applications is the field $\tilde{\phi}_\omega(\mathbf{x})$ at a position \mathbf{x} outside of a charge distribution. Setting the origin of the coordinate system equal to the center-of-mass of the charges one has $r = |\mathbf{x}| > |\mathbf{x}'| = r'$ and therefore $r_< = r', r_> = r$.

$$\begin{aligned} \Rightarrow \tilde{\phi}_\omega(\mathbf{x}) &= \frac{ik}{\varepsilon_0} \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}(\vartheta, \varphi) h_l^{(1)}(kr) \\ &\quad \cdot \int j_l(kr') Y_{lm}^*(\vartheta', \varphi') \tilde{\rho}_\omega(r', \vartheta', \varphi') d\Omega' dr' \end{aligned} \quad (1.17)$$

In many applications the extension of the charge distribution is small against the wave length $\lambda = \frac{2\pi}{k} = \frac{2\pi c}{\omega}$ (e.g. an atom or molecule: $r' = 1 \text{ \AA}$, $\lambda = 1 \mu\text{m}$). The argument kr' of j_l is therefore small. For $x \ll 1$ we have

$$\begin{aligned} j_l(x) &\approx \frac{x^l}{(2l+1)!!} \\ \Rightarrow \int j_l(kr') Y_{lm}^* \tilde{\rho}_\omega d\Omega' dr' &\approx \frac{k^l}{(2l+1)!!} \int r'^l Y_{lm}^* \tilde{\rho}_\omega d\Omega' dr' \end{aligned} \quad (1.18)$$

The spherical multipole moment of order lm is defined by

$$Q_{lm} := \int r'^l Y_{lm}^* \tilde{\rho}_\omega d\Omega' dr' \quad (1.19)$$

Special cases:

$$\begin{aligned} Q_{00} &= \frac{1}{\sqrt{4\pi}} \int \tilde{\rho}_\omega(\mathbf{x}') d^3x' \\ &= \frac{Q}{\sqrt{4\pi}} \quad Q = \text{total charge of ion/molecule at } \omega \\ Q_{11} &= -\sqrt{\frac{3}{8\pi}} \int \tilde{\rho}_\omega(\mathbf{x}') (x' - iy') d^3x' = -\sqrt{\frac{3}{8\pi}} (p_x - ip_y) \\ Q_{10} &= \sqrt{\frac{3}{4\pi}} \int \tilde{\rho}_\omega(\mathbf{x}') z' d^3x = \sqrt{\frac{3}{4\pi}} p_z \end{aligned}$$

where $\mathbf{p} = (p_x, p_y, p_z)$ is the (Cartesian) electric dipole moment:

$$\mathbf{p} = \int \mathbf{x}' \rho(\mathbf{x}') d^3x'$$

A point dipole creates a potential of the form

$$\phi_{\text{Dip}}(\mathbf{x}) = \frac{\mathbf{p} \cdot \mathbf{x}}{4\pi\epsilon_0 |\mathbf{x}|^3}$$

Physical meaning of a point dipole: to charges of equal magnitude but opposite sign at a very small distance L .

$$\text{Dipole moment} = q(L) \cdot L\mathbf{n} = \mathbf{p}$$

Let L go to 0 and $q(L) \rightarrow \infty$, in such a way that $q(L) \cdot L$ remains finite.

The electric field of a point dipole at the origin is given by

$$\mathbf{E}_{\text{Dip}}(\mathbf{x}) = \frac{1}{4\pi\epsilon_0} \frac{3\hat{\mathbf{x}}(\mathbf{p} \cdot \hat{\mathbf{x}}) - \mathbf{p}}{|\mathbf{x}|^3} \quad \text{with } \hat{\mathbf{x}} = \frac{\mathbf{x}}{|\mathbf{x}|}$$

The general multipole expansion of electrostatical fields is

$$\phi(\mathbf{x}) = \frac{1}{4\pi\epsilon_0} \left[\frac{Q}{r} + \frac{\mathbf{p} \cdot \mathbf{x}}{r^3} + \frac{1}{2} \sum_{ij} \bar{Q}_{ij} \frac{x_i x_j}{r^5} + \dots \right]$$

where

$$\bar{Q}_{ij} = \int \varrho(\mathbf{x}) (3x_i x_j - \delta_{ij} \mathbf{x}^2) d^3x$$

is the (traceless) tensor of the quadrupole moment.

Eq. (1.17) can also be derived by an expansion in spherical harmonics. They are eigenstates of the angular momentum operator and fulfill the orthogonality relation

$$\int_0^{2\pi} \int_0^\pi Y_{l'm'}^*(\vartheta, \varphi) Y_{lm}(\vartheta, \varphi) \sin \vartheta d\vartheta d\varphi = \delta_{ll'} \delta_{mm'} \quad (1.20)$$

and the completeness relation

$$\sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\vartheta', \varphi') Y_{lm}(\vartheta, \varphi) = \underbrace{\delta(\varphi - \varphi') \delta(\cos \vartheta - \cos \vartheta')}_{=\delta(\Omega - \Omega')} \quad (1.21)$$

Because of Eq. (1.21) we have for any scalar function $f(\vartheta, \varphi)$:

$$\begin{aligned} f(\vartheta, \varphi) &= \int f(\vartheta', \varphi') \delta(\Omega - \Omega') d\Omega' \\ &= \sum_{l,m} \int Y_{lm}^*(\Omega') Y_{lm}(\Omega) f(\Omega') d\Omega' \\ &= \sum_{l,m} Y_{lm}(\vartheta, \varphi) \cdot F_{lm} \quad \text{with coefficients } F_{lm} := \int Y_{lm}^*(\Omega') f(\Omega') d\Omega' \end{aligned}$$

In our case

$$\tilde{\phi}_\omega(\mathbf{x}) = \sum_{l,m} Y_{lm}(\vartheta, \varphi) \tilde{\phi}_{\omega,lm}(\mathbf{r})$$

$$\text{with } \tilde{\phi}_{\omega,lm}(\mathbf{r}) = \int Y_{lm}^*(\Omega') \tilde{\phi}_\omega(r', \vartheta', \varphi') d\Omega'$$

This can be inserted into the Helmholtz equation

$$(\Delta + k^2) \tilde{\phi}_\omega(\mathbf{x}) = -\frac{1}{\varepsilon_0} \tilde{\varrho}_\omega(\mathbf{x})$$

Laplace operator spherical coordinates:

$$\Delta = \frac{1}{r^2} \frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r} - \frac{1}{r^2} \mathbf{L}^2 \quad ; \quad \mathbf{L} = \mathbf{x} \times (-i\nabla)$$

$$\mathbf{L}^2 Y_{lm} = l(l+1) Y_{lm}$$

$$\Rightarrow \left(\frac{1}{r^2} \frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r} - \frac{l(l+1)}{r^2} + k^2 \right) \tilde{\phi}_{\omega,lm}(\mathbf{r}) = -\frac{1}{\varepsilon_0} \tilde{\varrho}_{\omega,lm}(\mathbf{r})$$

Solving this differential equation with suitable boundary conditions (outgoing radial wave for $r \rightarrow \infty$, i.e. $\sim \cos(kr)_r^{\frac{1}{2}} \sin(kr)_r^{\frac{1}{2}}$) leads to expression (1.17) for $\tilde{\phi}_\omega(\mathbf{x})$.

Multipole expansion for vector fields

It is also possible to make a multipole expansion of *divergence-free* vector fields (e.g., $\text{div} \mathbf{B} = 0$). One essentially replaces the spherical harmonics Y_{lm} by

$$\mathbf{X}_{lm} := \frac{1}{\sqrt{l(l+1)}} \mathbf{L} Y_{lm}(\vartheta, \varphi) \quad \text{''Vector spherical harmonics''}$$

\mathbf{X}_{lm} fullfills

$$\int \mathbf{X}_{lm}^* \mathbf{X}_{l'm'} d\Omega = \delta_{ll'} \delta_{mm'}$$

$$\int \mathbf{X}_{lm}^* \cdot (\mathbf{x} \times \mathbf{X}_{lm}) d\Omega = 0$$

A complete set of vector functions for divergence-free fields is then given by

$$F_l(kr) \mathbf{X}_{lm} \quad \text{and} \quad \nabla \times (g_l(kr) \mathbf{X}_{lm})$$

with $F_l(kr) = F_l^{(1)} h_l^{(1)}(kr) + F_l^{(2)} h_l^{(2)}(kr)$
 $g_l(kr) = g_l^{(1)} h_l^{(1)}(kr) + g_l^{(2)} h_l^{(2)}(kr)$
 $h_l^{(i)}(kr) = \text{spherical Hankel functions } h_l^{(2)}(kr) = h_l^{(1)*}(kr)$

The electromagnetic field in free space can then be expanded according to

$$\mathbf{B} = \sum_{l,m} [a_E(l, m) F_l(kr) \mathbf{X}_{lm} + a_m(l, m) \nabla \times g_l(kr) \mathbf{X}_{lm}]$$

$$\mathbf{E} = \sum_{l,m} \left[\frac{i}{k} a_E(l, m) \nabla \times F_l(kr) \mathbf{X}_{lm} + a_m(l, m) g_l(kr) \mathbf{X}_{lm} \right]$$

Fields proportional to a_m are called *spherical TM fields* ("transverse magnetic"), and fields proportional to a_E are called *spherical TE fields* ("transverse electrical").

These fields are associated to the corresponding multipole moments if $kr \ll 1$ is valid inside the charge distribution. One then finds

$$a_E(l, m) \sim Q_{lm} \quad ; \quad a_m(l, m) \sim M_{lm}$$

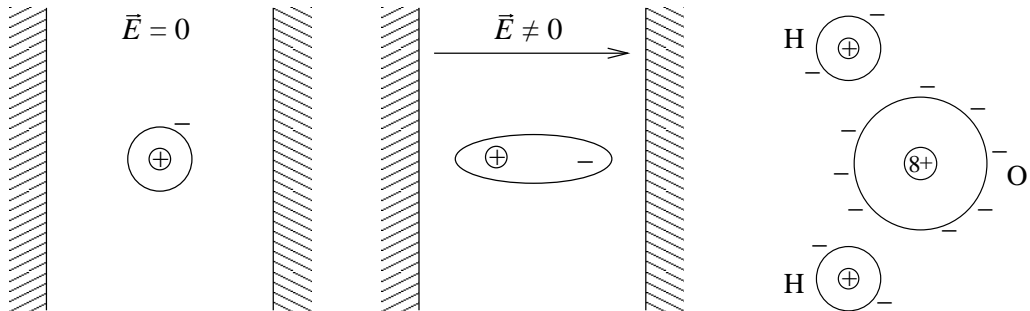
$$M_{lm} = -\frac{1}{(l+1)} \int r^l Y_{lm} \nabla \left(\frac{\mathbf{x} + \mathbf{j}(\mathbf{x})}{c} \right) d^3x$$

M_{lm} is called magnetic multipole moment. More on the vector multipole expansion can be found in Ref. [1], in particular in chapter 16.2 and 16.6.

1.5 Electrodynamics in Dielectric Media

Up to now we have only considered electromagnetic fields in free space in the presence of given charge or current distributions. However, one often wants to describe electromagnetic fields inside of some medium (e.g., glass or a crystal).

Almost all media consist of atoms and molecules. In such systems the charges are in a bound state, i.e., they cannot be substantially displaced by an applied external field. However, they are polarisable or even have a permanent dipole moment. Atoms, for instance, are polarisable by an external electric field:



Molecules (water, for instance) can also have a permanent dipole moment. This is often the case for ionic bounds. If one is only interested in describing macroscopic fields which are averaged over a volume containing many molecules, the evolution of the macroscopic fields \mathbf{E} , \mathbf{D} , \mathbf{B} and \mathbf{H} is governed by

$$\begin{aligned} \operatorname{div} \mathbf{D} &= \rho & \operatorname{div} \mathbf{B} &= 0 \\ \operatorname{curl} \mathbf{E} &= -\dot{\mathbf{B}} & \operatorname{curl} \mathbf{H} &= \mathbf{j} + \dot{\mathbf{D}} \end{aligned} \quad (1.22)$$

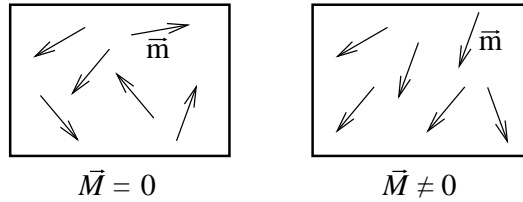
macroscopic Maxwell equations

The influence of the specific medium is contained in the relations

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \quad ; \quad \mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M})$$

Here \mathbf{D} is the dielectric displacement and \mathbf{H} the magnetic field, \mathbf{P} is the polarisation, and \mathbf{M} magnetisation of the medium. There are several special cases:

- In a vacuum we have $\mathbf{P} = 0$ and $\mathbf{M} = 0$, leading to the ordinary Maxwell equations.
- If molecules have a permanent (electric or magnetic) dipole moment but are randomly aligned inside the medium, one still finds $\mathbf{M} = \mathbf{P} = 0$ because of the averaging. If one aligns the molecules \mathbf{M} or $\mathbf{P} \neq 0$ is possible. The classical example for a model of this behaviour is the Ising model.



- If the atoms/molecules are polarisable, one finds

$$\mathbf{P} = \chi \mathbf{E} \quad ; \quad \mathbf{D} = \varepsilon \mathbf{E} = \varepsilon_0 \varepsilon_r \mathbf{E} = (\varepsilon + \chi) \mathbf{E}$$

ε is the dielectric constant of the medium and χ its susceptibility. This effect can (particularly in crystals) depend on the direction of the fields (birefringence):

$$P_i = \chi_{ij} E_j \quad ; \quad D_i = \varepsilon_{ij} E_j$$

ε_{ij} : dielectric tensor.

- According to the model of Fedorov optically active media are described by²

$$\mathbf{M} = 0 \quad ; \quad \mathbf{D} = \varepsilon(\mathbf{E} + \beta \operatorname{curl} \mathbf{E})$$

The meaning of optical activity is that the polarization of a light beams rotates during the propagation through the medium.

- In nonlinear media one has, for instance,

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \kappa |\mathbf{E}|^2 \mathbf{E}$$

1.6 Derivation of macroscopic Maxwell equations

The fields \mathbf{E} , \mathbf{D} , \mathbf{B} , and \mathbf{H} describe macroscopic fields which result from an averaging over "microscopic" fields \mathbf{E}_{mic} and \mathbf{B}_{mic} . The latter fullfill the usual Maxwell equations

$$\begin{aligned} \operatorname{div} \mathbf{E}_{\text{mic}} &= \frac{\varrho_{\text{mic}}}{\varepsilon_0} & \operatorname{div} \mathbf{B}_{\text{mic}} &= 0 \\ \operatorname{curl} \mathbf{E}_{\text{mic}} &= -\dot{\mathbf{B}}_{\text{mic}} & \operatorname{curl} \mathbf{B}_{\text{mic}} &= \mu_0 (\mathbf{j}_{\text{mic}} + \varepsilon_0 \dot{\mathbf{E}}_{\text{mic}}) \end{aligned}$$

Charge and current distributions are divided into two parts,

$$\begin{aligned} \varrho_{\text{mic}} &= \varrho_{\text{free}} + \varrho_{\text{bound}} \\ \mathbf{j}_{\text{mic}} &= \mathbf{j}_{\text{free}} + \mathbf{j}_{\text{bound}} \end{aligned}$$

The free quantities describe particles which are not bound to atoms/molecules. In classical mechanics their charge distribution can be written as

$$\begin{aligned} \varrho_{\text{free}}(\mathbf{x}) &= \sum_i q_i \delta(\mathbf{x} - \mathbf{x}_i) \\ \mathbf{j}_{\text{free}}(\mathbf{x}) &= \sum_i q_i \dot{\mathbf{x}}_i \delta(\mathbf{x} - \mathbf{x}_i) \quad \text{Sum over free particles.} \end{aligned}$$

The bound quantities correspond to the atoms and molecules of the medium.

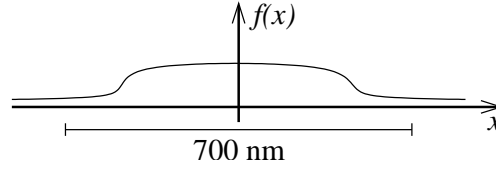
$$\begin{aligned} \varrho_{\text{bound}}(\mathbf{x}) &= \sum_n \varrho_n(\mathbf{x}) \quad \text{Sum over molecules} \\ \varrho_n(\mathbf{x}) &= \sum_i q_i \delta(\mathbf{x} - \mathbf{x}_i) \quad \text{Sum over the particles } (e^-, \text{ nuclei}) \text{ of the molecule} \\ \mathbf{j}_{\text{bound}}(\mathbf{x}) &= \sum_n \mathbf{j}_n(\mathbf{x}) \quad ; \quad \mathbf{j}_n(\mathbf{x}) = \sum_i q_i \dot{\mathbf{x}}_i \delta(\mathbf{x} - \mathbf{x}_i) \end{aligned}$$

²There are several models for optical activity. We use Fedorov's model just as an example.

In most situations experimental measurements cannot resolve electromagnetic fields of individual molecules (size $\sim 1 \text{ \AA}$). In optical experiments the spatial resolution is in the order of a wavelength ($\sim 6000 \text{ \AA}$). One therefore can perform a spatial averaging over \mathbf{E}_{mic} and \mathbf{B}_{mic} without getting a worse description of the experiments.

$$\begin{aligned}\mathbf{E}(\mathbf{x}) &= \langle \mathbf{E}_{\text{mic}}(\mathbf{x}) \rangle = \int f(\mathbf{x}') \mathbf{E}_{\text{mic}}(\mathbf{x} - \mathbf{x}') d^3x' \\ \mathbf{B}(\mathbf{x}) &= \langle \mathbf{B}_{\text{mic}}(\mathbf{x}) \rangle\end{aligned}$$

In these expressions $f(\mathbf{x})$ is a function which varies slowly on molecular scales, but the support of which is small against the wavelength. Because we consider an averaging procedure we of course demand $\int f(\mathbf{x}) d^3x = 1$.



The equations for \mathbf{E} and \mathbf{B} then become

$$\begin{aligned}\text{div} \mathbf{B}(\mathbf{x}) &= \frac{\partial}{\partial x_i} B_i(\mathbf{x}) \\ &= \frac{\partial}{\partial x_i} \int f(\mathbf{x}') B_{\text{mic},i}(\mathbf{x} - \mathbf{x}') d^3x' \\ &= \int f(\mathbf{x}') \frac{\partial}{\partial x_i} B_{\text{mic},i}(\mathbf{x} - \mathbf{x}') d^3x' \\ &= \int f(\mathbf{x}') \underbrace{\text{div} \mathbf{B}_{\text{mic}}(\mathbf{x} - \mathbf{x}')}_{=0} d^3x' \\ &= \langle \text{div} \mathbf{B}_{\text{mic}}(\mathbf{x}) \rangle = 0\end{aligned}$$

$$\begin{aligned}\text{and analogously } \text{div} \mathbf{E}(\mathbf{x}) &= \frac{\langle \varrho_{\text{mic}} \rangle}{\varepsilon_0} \\ \text{curl} \mathbf{E}(\mathbf{x}) &= -\dot{\mathbf{B}}(\mathbf{x}) \\ \text{curl} \mathbf{B}(\mathbf{x}) &= \mu_0 \langle \mathbf{j}_{\text{mic}}(\mathbf{x}) \rangle + \frac{1}{c^2} \dot{\mathbf{E}}(\mathbf{x})\end{aligned}$$

The mean values can be calculated as follows.

$$\begin{aligned}\langle \varrho_{\text{mic}} \rangle &= \langle \varrho_{\text{free}} \rangle + \langle \varrho_{\text{bound}} \rangle \\ &=: \varrho(\mathbf{x}) + \langle \varrho_{\text{bound}} \rangle \quad \varrho(\mathbf{x}) : \text{macroscopic charge density} \\ \langle \varrho_{\text{bound}} \rangle &= \sum_{n(\text{molecules})} \langle \varrho_n(\mathbf{x}) \rangle\end{aligned} \tag{1.23}$$

The size of each molecule is much smaller than the averaging area. We therefore find for molecule n

$$\begin{aligned}\langle \varrho_n(\mathbf{x}) \rangle &= \int f(\mathbf{x}') \varrho_n(\mathbf{x} - \mathbf{x}') d^3x' \\ &= \int f(\mathbf{x}') \sum_{i(e^-, \text{nuclei})} q_i \delta(\mathbf{x} - \mathbf{x}' - \mathbf{x}_i) d^3x' \\ &= \sum_{i(e^-, \text{nuclei})} q_i f(\mathbf{x} - \mathbf{x}_i)\end{aligned}$$

If \mathbf{x}_n is the center of the molecule we have $|\mathbf{x}_i - \mathbf{x}_n| \ll \text{support of } f$. One therefore can perform a Taylor expansion of f :

$$\begin{aligned}f(\mathbf{x} - \mathbf{x}_i) &= f(\mathbf{x} - \mathbf{x}_n - (\mathbf{x}_i - \mathbf{x}_n)) \\ &\approx f(\mathbf{x} - \mathbf{x}_n) - (\mathbf{x}_i - \mathbf{x}_n) \cdot \nabla f(\mathbf{x} - \mathbf{x}_n) + \dots \\ \Rightarrow \langle \varrho_n(\mathbf{x}) \rangle &\approx \sum_{i(e^-, \text{nuclei})} q_i \{f(\mathbf{x} - \mathbf{x}_n) - (\mathbf{x}_i - \mathbf{x}_n) \cdot \nabla f(\mathbf{x} - \mathbf{x}_n) + \dots\} \\ \Rightarrow \langle \varrho_n(\mathbf{x}) \rangle &\approx f(\mathbf{x} - \mathbf{x}_n) \underbrace{\sum_{i(e^-, \text{nuclei})} q_i}_{q_n} - \nabla f(\mathbf{x} - \mathbf{x}_n) \cdot \underbrace{\sum_{i(e^-, \text{nuclei})} q_i (\mathbf{x}_i - \mathbf{x}_n)}_{\mathbf{p}_n} + \dots\end{aligned}$$

q_n is the total charge of the n th molecule. If the molecules are not ionized, $q_n = 0$. \mathbf{p}_n is the molecule's dipole moment.

$$\Rightarrow \langle \varrho_{\text{bound}} \rangle = - \sum_{n(\text{molecules})} \mathbf{p}_n \cdot \nabla f(\mathbf{x} - \mathbf{x}_n)$$

Define the macroscopic polarization $\mathbf{P}(\mathbf{x}) = \sum_n f(\mathbf{x} - \mathbf{x}_n) \mathbf{p}_n$; $q_n = 0$

$$\Rightarrow \boxed{\langle \varrho_{\text{bound}}(\mathbf{x}) \rangle = -\text{div} \mathbf{P}(\mathbf{x})}$$

Inserting this and Eq. (1.23) into the microscopic Maxwell equations one gets

$$\begin{aligned}\text{div} \mathbf{E} &= \frac{1}{\varepsilon_0} \langle \varrho_{\text{mic}} \rangle \\ &= \frac{1}{\varepsilon_0} \varrho - \frac{1}{\varepsilon_0} \text{div} \mathbf{P} \\ \mathbf{D} &= \varepsilon_0 \mathbf{E} + \mathbf{P} \Rightarrow \text{div} \mathbf{D} = \varrho\end{aligned}$$

A similar treatment of

$$\text{curl} \mathbf{B} = \mu_0 \langle \mathbf{j}_{\text{mic}} \rangle + \frac{1}{c^2} \dot{\mathbf{E}}$$

leads to

$$\begin{aligned}\langle \dot{\mathbf{j}}_{\text{mic}} \rangle &= \dot{\mathbf{j}} + \text{curl } \mathbf{M} + \dot{\mathbf{P}} \\ \mathbf{M} &= \left\langle \sum_n \mathbf{m}_n \delta(\mathbf{x} - \mathbf{x}_n) \right\rangle \\ \mathbf{m}_n &= \sum_{i(e^-, \text{nuclei})} \frac{q_i}{2m_i} (\mathbf{x}_i \times m_i \dot{\mathbf{x}}_i) = \sum_{i(e^-, \text{nuclei})} \frac{q_i}{2m_i} \mathbf{L}_i\end{aligned}$$

Here \mathbf{m}_n denotes the magnetic dipole moment of the n th molecule. (details can be found in [1], section 6.7). This concludes the derivation of the macroscopic Maxwell equations Eq. (1.22).

1.7 Plane Waves

In vacuum ($\varrho = \mathbf{j} = 0$) the following relations hold,

$$\square \mathbf{E} = \square \mathbf{B} = 0 \quad ; \quad \text{div } \mathbf{E} = \text{div } \mathbf{B} = 0$$

Ansatz for solution:

$$\begin{aligned}\mathbf{E} &= \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}_{\mathbf{k}} \exp(i\mathbf{k}\mathbf{x} - i\omega t) \\ \square \mathbf{E} &= \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}_{\mathbf{k}} \left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right) \exp(i\mathbf{k}\mathbf{x} - i\omega t)\end{aligned}$$

where $\mathcal{E}_{\mathbf{k}} \in \mathbb{C}$ is the amplitude and $\boldsymbol{\epsilon}_{\mathbf{k}}$ the polarisation vector.

$$\implies \square \mathbf{E} = \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}_{\mathbf{k}} \left(-\frac{\omega^2}{c^2} + \mathbf{k}^2 \right) \exp(i\mathbf{k}\mathbf{x} - i\omega t) \stackrel{!}{=} 0$$

This is a solution of $\square \mathbf{E} = 0$ if $\omega = c|\mathbf{k}|$, where $\omega_{\mathbf{k}} := c|\mathbf{k}|$ is the frequency of a photon³ with momentum $\hbar\mathbf{k}$. Its energy is given by Planck's equation $E = \hbar\omega_{\mathbf{k}}$. The polarisation vector $\boldsymbol{\epsilon}_{\mathbf{k}}$ is determined by the condition $\text{div } \mathbf{E} = \partial_i E_i = 0$:

$$\begin{aligned}\text{div } \mathbf{E} &= \mathcal{E}_{\mathbf{k}} (\boldsymbol{\epsilon}_{\mathbf{k}})_i i k_i \exp(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}} t) \\ &= i \mathcal{E}_{\mathbf{k}} (\boldsymbol{\epsilon}_{\mathbf{k}} \mathbf{k}) \exp(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}} t) \\ &\stackrel{!}{=} 0\end{aligned}$$

³The notion of a photon as the elementary quantum of the radiation fields is introduced in Quantum Electrodynamics (QED). However, the properties of a photon are closely related to those of a classical electrodynamical field. We therefore use this notion already here.

$\Rightarrow \epsilon_{\mathbf{k}}$ must be perpendicular to the wave vector \mathbf{k} !

In \mathbb{R}^3 there are two linearly independent vectors perpendicular to $\mathbf{k} \Rightarrow$ there are two independent polarization vectors. The specific direction of $\epsilon_{\mathbf{k}}^{(\sigma)}$ is arbitrary. One possibility is

$$\mathbf{k} = k \begin{pmatrix} \cos \varphi \sin \vartheta \\ \sin \varphi \sin \vartheta \\ \cos \vartheta \end{pmatrix} \quad ; \quad \epsilon_{\mathbf{k}}^{(1)} = \begin{pmatrix} \cos \varphi \cos \vartheta \\ \sin \varphi \cos \vartheta \\ -\sin \vartheta \end{pmatrix} \quad ; \quad \epsilon_{\mathbf{k}}^{(2)} = \frac{\mathbf{k}}{k} \times \epsilon_{\mathbf{k}}^{(1)} = \begin{pmatrix} -\sin \varphi \\ \cos \varphi \\ 0 \end{pmatrix}$$

The real vectors $\epsilon_{\mathbf{k}}^{(1)}$ and $\epsilon_{\mathbf{k}}^{(2)}$ correspond to linearly polarized light. Generally the polarization is a (complex) superposition of these two vectors,

$$\epsilon_{\mathbf{k}} = \alpha \epsilon_{\mathbf{k}}^{(1)} + \beta \epsilon_{\mathbf{k}}^{(2)} \quad ; \quad \text{with } |\alpha|^2 + |\beta|^2 = 1 \quad , \quad \alpha, \beta \in \mathbb{C}$$

An example is circularly polarized light:

$$\epsilon_{\mathbf{k}}^{(\pm)} = \frac{1}{\sqrt{2}} (\epsilon_{\mathbf{k}}^{(1)} \pm i \epsilon_{\mathbf{k}}^{(2)})$$

Different conventions are used in the literature for the names of circular light beams. We use that of Ref. [1, 11]:

”+”: left circular or positive helicity, σ^+ light

”-”: right circular or negative helicity, σ^- light

σ^+ - and σ^- light plays an important role in selection rules for atomic transitions: $\sigma^\pm : \Delta m = \pm 1$, where m is the magnetic quantum number of the atomic state. General superposition vectors $\epsilon_{\mathbf{k}} = \alpha \epsilon_{\mathbf{k}}^{(1)} + \beta \epsilon_{\mathbf{k}}^{(2)}$ are called elliptically polarized.

In Quantum Electrodynamics the polarization vector is connected to the spin of a photon. Photons have total spin 1, but their spin in z -direction can only take the values $s_z = \pm 1$. This corresponds to σ^+ - and σ^- light. The selection rule $\Delta m = \pm 1$ is then simply a consequence of angular momentum conservation when the atom absorbs a photon. The value $s_z = 0$ is not realized in nature because of the vanishing rest mass of photons (see, e.g., Ref. [9], section 14.5).

The magnetic field of a plane wave can be derived from the electric field with the aid of the Maxwell equation $\text{curl } \mathbf{E} = -\dot{\mathbf{B}}$:

$$\begin{aligned} \dot{\mathbf{B}} &= -\text{curl} \{ \mathcal{E}_{\mathbf{k}} \epsilon_{\mathbf{k}} e^{(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}}t)} + \text{c.c.} \} \\ &= -\epsilon_{ijk} \partial_j (\epsilon_{\mathbf{k}})_l \mathcal{E}_{\mathbf{k}} e^{(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}}t)} + \text{c.c.} \\ &= -\epsilon_{ijk} (\epsilon_{\mathbf{k}})_l \mathcal{E}_{\mathbf{k}} l k_j e^{(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}}t)} + \text{c.c.} \\ &= -i \mathcal{E}_{\mathbf{k}} (\mathbf{k} \times \epsilon_{\mathbf{k}})_i e^{(i\mathbf{k}\mathbf{x} - i\omega_{\mathbf{k}}t)} + \text{c.c.} \end{aligned}$$

To solve this we set $B_i \sim \exp(-i\omega_k t)$. We then find

$$\begin{aligned} \mathbf{B} &= \frac{\mathcal{E}_{\mathbf{k}}}{\omega_k} (\mathbf{k} \times \boldsymbol{\epsilon}_{\mathbf{k}}) e^{(i\mathbf{k}\mathbf{x} - i\omega_k t)} + \text{c.c.} \\ &= \frac{1}{\omega_k} (\mathbf{k} \times \mathbf{E}) \end{aligned}$$

The general solution in free space is a superposition of plane waves

$$\begin{aligned} \mathbf{E}(\mathbf{x}, t) &= \int d^3k \sum_{\sigma=1,2} \mathcal{E}_{\mathbf{k}}^{(\sigma)} \boldsymbol{\epsilon}_{\mathbf{k}}^{(\sigma)} \exp(i\mathbf{k}\mathbf{x} - i\omega t) + \text{c.c.} \\ \mathbf{B}(\mathbf{x}, t) &= \int d^3k \sum_{\sigma=1,2} \frac{\mathcal{E}_{\mathbf{k}}^{(\sigma)}}{\omega_k} (\mathbf{k} \times \boldsymbol{\epsilon}_{\mathbf{k}}^{(\sigma)}) \exp(i\mathbf{k}\mathbf{x} - i\omega t) + \text{c.c.} \end{aligned}$$

The following special cases are worth to be mentioned:

1. The polarization vector is composed of a real part $\boldsymbol{\epsilon}$ and a imaginary part $\boldsymbol{\epsilon}''$,

$$\begin{aligned} \mathbf{E} &= \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}_{\mathbf{k}} \exp(i\mathbf{k}\mathbf{x} - i\omega_k t) + \text{c.c.} \\ &= \mathcal{E}_{\mathbf{k}} (\boldsymbol{\epsilon}' + i\boldsymbol{\epsilon}'') \exp(i\mathbf{k}\mathbf{x} - i\omega_k t) + \text{c.c.} \end{aligned}$$

where we assume $\boldsymbol{\epsilon}'^2 + \boldsymbol{\epsilon}''^2 = 1$, $\mathcal{E} \in \mathbb{R}$.

$$\implies \mathbf{E} = \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}' 2 \cos(\mathbf{k}\mathbf{x} - i\omega_k t) - \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}'' 2 \sin(\mathbf{k}\mathbf{x} - i\omega_k t)$$

This is a running wave.

2. Consider a superposition of two counter-propagating waves with $\mathcal{E}_{\mathbf{k}} = \mathcal{E}_{-\mathbf{k}}$ and $\boldsymbol{\epsilon}_{\mathbf{k}} = \boldsymbol{\epsilon}_{-\mathbf{k}}$:

$$\begin{aligned} \mathbf{E} &= \mathbf{E}_{\mathbf{k}} + \mathbf{E}_{-\mathbf{k}} \\ &= \mathcal{E}_{\mathbf{k}} \boldsymbol{\epsilon}_{\mathbf{k}} \exp(i\mathbf{k}\mathbf{x} - i\omega_k t) + \mathcal{E}_{-\mathbf{k}} \boldsymbol{\epsilon}_{-\mathbf{k}} \exp(-i\mathbf{k}\mathbf{x} - i\omega_k t) + \text{c.c.} \\ &\implies \mathbf{E} \sim \cos(\mathbf{k}\mathbf{x}) \cos(\omega_k t) \end{aligned}$$

This is a standing wave.

1.8 Paraxial approximation, focused light beams

The paraxial approximation allows to describe the shape of a laser beam or the propagation of a laser pulse in a much simpler way. It reduces the second-order wave equation to a first-order differential equation along the axis of propagation.

As we will see in Chapter 8, it can also be used to calculate the optical properties of a medium.

The idea behind the paraxial approximation is to consider a light field which is a running wave with a spatially (and/or temporally) varying envelope $\mathcal{E}(\mathbf{x}, t)$. The variation of the envelope should be small on the scales of one wavelength (and/or one optical period of time). We write the field as

$$\mathbf{E} = \epsilon \mathcal{E}(\mathbf{x}, t) e^{-i\omega t} e^{ikz} \quad ; \quad k = \frac{\omega}{c} \quad (\text{beam in } z\text{-direction}) \quad (1.24)$$

and consider (here) solutions of the wave equation in free space. It follows that

$$\begin{aligned} 0 &= \square \mathbf{E} \\ &= \left(\frac{1}{c^2} \partial_t^2 - \Delta \right) \mathcal{E}(\mathbf{x}, t) e^{ikz - i\omega t} \epsilon \\ &= \left\{ \frac{1}{c^2} (\partial_t^2 \mathcal{E} - 2i\omega \partial_t \mathcal{E} - \omega^2 \mathcal{E}) - (\Delta_{\perp} \mathcal{E} + \partial_z^2 \mathcal{E} + 2ik \partial_z \mathcal{E} - k^2 \mathcal{E}) \right\} e^{ikz - i\omega t} \epsilon \\ &= \left\{ \frac{1}{c^2} \partial_t^2 \mathcal{E} - 2ik \frac{1}{c} \partial_t \mathcal{E} - \Delta_{\perp} \mathcal{E} - \partial_z^2 \mathcal{E} - 2ik \partial_z \mathcal{E} \right\} e^{ikz - i\omega t} \epsilon \\ &= \left\{ \left(-2ik \left(\frac{1}{c} \partial_t + \partial_z \right) - \Delta_{\perp} \right) \mathcal{E} + \frac{1}{c^2} \partial_t^2 \mathcal{E} - \partial_z^2 \mathcal{E} \right\} e^{ikz - i\omega t} \epsilon \end{aligned} \quad (1.25)$$

with $\Delta_{\perp} \equiv \partial_x^2 + \partial_y^2$. We assume that the envelope $\mathcal{E}(\mathbf{x}, t)$ varies slowly with z compared to e^{ikz} , and with t compared to $e^{-i\omega t}$. This implies that

$$k \partial_z \mathcal{E} \gg \partial_z^2 \mathcal{E} \quad , \quad \frac{k}{c} \partial_t \mathcal{E} \gg \partial_t^2 \mathcal{E} . \quad (1.26)$$

We then can neglect the second derivatives with respect to z and t and are led to the *paraxial wave equation*

$$\boxed{2ik \left(\frac{1}{c} \partial_t + \partial_z \right) \mathcal{E} = -\Delta_{\perp} \mathcal{E} .} \quad (1.27)$$

When the light field is monochromatic the envelope is constant in time and we arrive at the Paraxial Helmholtz equation

$$\boxed{2ik \partial_z \mathcal{E} = -\Delta_{\perp} \mathcal{E} .}$$

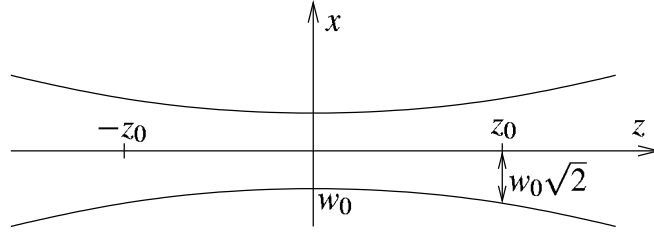
This equation has the same structure as the free Schrödinger equation in two dimensions ($z \hat{=} t$).

We know how to solve this equations from quantum mechanics. If the solution has a transverse Gaussian profile of width w_0 at $z = 0$, the envelope of the laser beam is given by

$$\mathcal{E}(z, \varrho) = E_{\max} \frac{z_R}{q(z)} e^{ik \frac{\varrho^2}{2q(z)}} \quad \text{with } q(z) = z - iz_R \text{ and } \varrho = \sqrt{x^2 + y^2}$$

The quantity $z_R \equiv kw_0^2$ is called Rayleigh length. The intensity of the beam is proportional to $|\mathbf{E}|^2$:

$$I = I_0 \left(\frac{w_0}{w(z)} \right)^2 e^{-\frac{r^2}{w^2(z)}} \quad , \quad w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R} \right)^2}$$

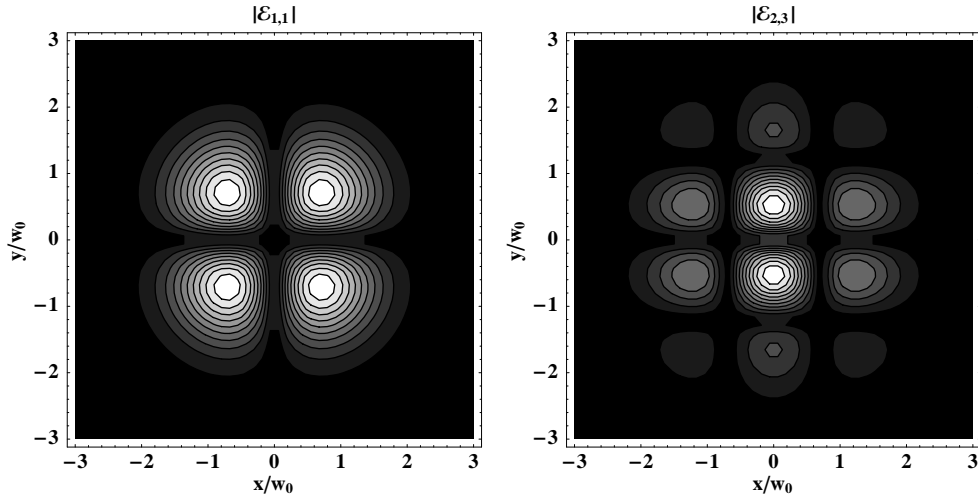


The Rayleigh length is connected to the degree of focusing (the minimal width) w_0 of the beam. Its physical interpretation is that it corresponds to the length over which the area of the light beam increases by a factor of two. The smaller z_R is, the smaller is also the minimal width w_0 , but also the range over which the beam is strongly focused.

Other important solutions (Laser modes) are the Hermite-Gaussian beams:

$$\mathcal{E}_{l,m} = H_l(x/\sqrt{a}) H_m(y/\sqrt{a}) e^{-\frac{x^2+y^2}{a}} e^{-\frac{l+m+2}{2} \ln(a/w_0^2)} \quad (1.28)$$

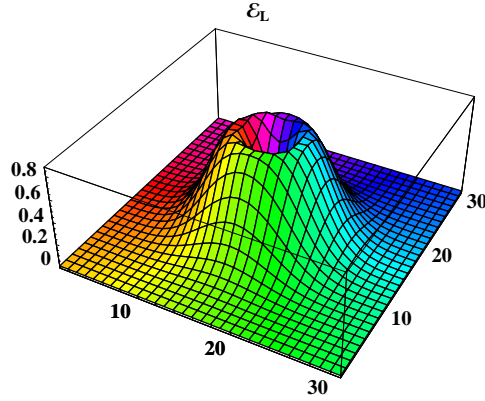
with $a(z) \equiv w_0^2(1 + 2iz/z_R)$ and H_l the Hermite polynomials.



Of particular interest are complex superpositions of these modes. For instance, the superposition $\mathcal{E}_L = \mathcal{E}_{1,0} + i\mathcal{E}_{0,1}$ takes the form

$$\mathcal{E}_L = e^{ik\frac{r^2}{2q(z)}} \frac{(x + iy)z_R}{(2iz + z_R)^2} \quad (1.29)$$

The presence of the factor $x + iy$ indicates that light in this mode possesses an orbital angular momentum. In the figure below this is indicated by the color change which represents the variation of the phase of the field (red corresponds to zero, the colors then circle through the rainbow colors).



Formally it can be shown (see, e.g., Ref. [9], Sec. 14.3) that the angular momentum operator for electromagnetic field is given by

$$M_{ij} = \hbar \int d^3x \dot{A}_r (x_i \partial_j - x_j \partial_i) A_r - \dot{A}_i A_j + \dot{A}_j A_i \quad (1.30)$$

It is easy to check that \mathcal{E}_L has an orbital angular momentum of $M_{12} = \hbar$ along its propagation axis. Such a field can be generated using specially shaped phase plates. An interesting application of these fields is that in the interaction between atoms and light not only energy and momentum but also angular momentum is conserved. One therefore can transfer orbital angular momentum from light to atoms. This effect has been used to generate vortices in Bose-Einstein condensates [46, 47].

Chapter 2

Quantum Field Theory of Light - QED

2.1 Many-Particle Theory of Quantum Mechanics

We assume that the reader is familiar with the Schrödinger equation for a single particle. If one considers two non-interacting free particles, the Hamiltonian is given by the sum of the individual Hamiltonians,

$$H = \frac{p_1^2}{2m} + \frac{p_2^2}{2m} \quad (2.1)$$

The corresponding two-particle wavefunction has the form

$$\psi(\mathbf{x}_1, \mathbf{x}_2) = \phi_n(\mathbf{x}_1) \cdot \phi_m(\mathbf{x}_2) \quad (2.2)$$

In this wavefunction the particles are distinguishable (through their quantum numbers n, m). However, experiments demonstrate that identical particles are NOT distinguishable. In the theoretical description this is guaranteed if $\psi(\mathbf{x}_1, \mathbf{x}_2)$ leads to the same measurement results as $\psi(\mathbf{x}_2, \mathbf{x}_1)$. To examine this more closely we introduce the exchange operator \hat{E} ,

$$\hat{E}\psi(\mathbf{x}_1, \mathbf{x}_2) = \psi(\mathbf{x}_2, \mathbf{x}_1) \quad (2.3)$$

Because of $\hat{E}^2 = \mathbb{1}$ the eigenvalues of the exchange operator are ± 1 . The corresponding eigenfunctions fulfill

$$\hat{E}\psi_B(\mathbf{x}_1, \mathbf{x}_2) = +\psi_B(\mathbf{x}_1, \mathbf{x}_2) = \psi_B(\mathbf{x}_2, \mathbf{x}_1) \quad (2.4)$$

$$\hat{E}\psi_F(\mathbf{x}_1, \mathbf{x}_2) = -\psi_F(\mathbf{x}_1, \mathbf{x}_2) = \psi_F(\mathbf{x}_2, \mathbf{x}_1) \quad (2.5)$$

All experimental results indicate that all particles in nature are eigenstates of \hat{E} :

- $\psi_B \Leftrightarrow$ Bosons
- $\psi_F \Leftrightarrow$ Fermions

The well-known Pauli principle for Fermions is a direct consequence of the anti-symmetry of ψ_F .

Spin-Statistics Theorem

- Bosons \Leftrightarrow interger spin
- Fermions \Leftrightarrow half-integer spin

The proof of this theorem essentially relies on causality and can be found in Ref. [5].

Many-particle Wavefunction $\psi(x_1, \dots, x_n)$ should be symmetric or antisymmetric under exchange of any pair of particles:

$$\psi(x_1, \dots, x_i, \dots, x_j, \dots, x_n) = \pm \psi(x_1, \dots, x_j, \dots, x_i, \dots, x_n) \quad (2.6)$$

This can be achieved by symmetrization: sum over all permutations and normalize

$$\psi_B(x_1, \dots, x_n) = \frac{1}{\sqrt{N!}} \sum_p \psi(x_{p(1)}, \dots, x_{p(n)}) \quad (2.7)$$

$$\psi_F(x_1, \dots, x_n) = \frac{1}{\sqrt{N!}} \sum_p (-1)^p \psi(x_{p(1)}, \dots, x_{p(n)}) \quad (2.8)$$

whereby p characterizes the permutation: $(-1)^p = 1$ for an even number of exchanges, and -1 for an odd number of exchanges.

As an example we use the product wavefunction of free particles,

$$\psi(x_1, x_2, x_3) = \psi_l(x_1)\psi_m(x_2)\psi_n(x_3) , \quad (2.9)$$

i.e., particle 1 in state l, particle 2 in state m, and particle 3 in state n. This means that the particle are distinguishable.

For Fermions the corresponding indistinguishable wavefunction is

$$\begin{aligned} \psi_{lmn}^F(x_1, x_2, x_3) = & \frac{1}{\sqrt{6}} \{ \psi_l(x_1)\psi_m(x_2)\psi_n(x_3) - \psi_l(x_2)\psi_m(x_1)\psi_n(x_3) \\ & + \psi_l(x_2)\psi_m(x_3)\psi_n(x_1) - \psi_l(x_3)\psi_m(x_2)\psi_n(x_1) \\ & + \psi_l(x_3)\psi_m(x_1)\psi_n(x_2) - \psi_l(x_1)\psi_m(x_3)\psi_n(x_2) \} \end{aligned}$$

For a large number of particles this procedure becomes rather tedious.

2.1.1 Fock Space and Number Representation

In single-particle quantum mechanics we usually do not need to know the full wavefunction $\psi(x)$ in position space. To completely fix the state it is sufficient to know a complete set of quantum numbers. For the hydrogen atom, for instance, these are the quantum numbers n, l and m (ignoring spin). To know the state it is sufficient to specify $|n, l, m\rangle$ instead of $\psi_{nlm}(x) = \langle x|n, l, m\rangle$.

Likewise, in many-particle quantum mechanics it is sufficient to know, how many particles are in a state: the ket $|1_l, 1_m, 1_n\rangle$ can unambiguously be associated with the state "one particle in state l , one in m , one in n ", i.e., with the wavefunction $\psi_{lmn}^F(x_1, x_2, x_3)$.

Example for Bosons:

$$\begin{aligned} \psi_{lmn}^B(x_1, x_2, x_3) &= \frac{1}{\sqrt{3}} \{ \psi_l(x_1)\psi_l(x_2)\psi_m(x_3) \\ &\quad + \psi_l(x_1)\psi_l(x_3)\psi_m(x_2) \\ &\quad + \psi_l(x_2)\psi_l(x_3)\psi_m(x_1) \} \\ &\Longleftrightarrow |2_l, 1_m\rangle \end{aligned}$$

Superposition of such states are possible and can also be prepared in an experiment. Example

$$|\psi\rangle = \alpha |2_l, 1_m\rangle + \beta |3_l, 0_m\rangle + \gamma |1_l, 2_m\rangle$$

However, also superpositions of states with a different number of particles, like

$$|\psi\rangle = \alpha |1_l\rangle + \beta |2_l\rangle + \gamma |3_l, 4_m\rangle \dots,$$

are possible. Formally superpositions of states with different particle numbers are an element of Fock space, which is defined as follows:

- Let $\mathcal{H}^{(1)}$ be the Hilbert space for 1-particle wavefunctions
- $\mathcal{H}^{(2)}$ = Hilbert space for 2-particle wavefunctions ...

Fock space is the direct sum of all N -particle Hilbert spaces:

$$F := \mathcal{H}^{(0)} \oplus \mathcal{H}^{(1)} \oplus \mathcal{H}^{(2)} \oplus \dots \oplus \mathcal{H}^{(N)} \quad (2.10)$$

$\mathcal{H}^{(0)}$ describes the vacuum and correspondingly includes only a single state, denoted by $|0\rangle$ ($\langle 0|0\rangle = 1$).

Fock space is needed to describe processes in which the number of particles is not conserved. For instance, pair production of e^+ and e^- from a γ photon is described by ¹

$$|1_\gamma\rangle \otimes |0_{e+}\rangle |0_{e-}\rangle \longrightarrow |0_\gamma\rangle \otimes |1_{e+}\rangle |1_{e-}\rangle$$

A second example is the excitation of one atom from the ground state $|1_g\rangle$ to the excited state $|1_e\rangle$ by absorbing a photon:

$$|1_g\rangle \otimes |1_\gamma\rangle \longrightarrow |1_e\rangle \otimes |0_\gamma\rangle$$

2.1.2 Creation and Anihilation Operators

These two types of operators allow a simple representation of many-particle wavefunctions. The creation operator a_l^\dagger for a particle in state l (also called mode l) and the corresponding anihilation operator a_l are defined by

$$a_l^\dagger |n_l^{(l)}, n_m^{(m)} \dots\rangle = \sqrt{n_l^{(l)} + 1} |(n_l^{(l)} + 1)_l, n_m^{(m)} \dots\rangle \quad (2.11)$$

$$a_l |n_l^{(l)}, n_m^{(m)} \dots\rangle = \sqrt{n_l^{(l)}} |(n_l^{(l)} - 1)_l, n_m^{(m)} \dots\rangle \quad (2.12)$$

These are the definitions for Bosons.

These operators fulfill the following commutation relations (For simplicity we omit the indices).

$$\begin{aligned} [a^\dagger, a] |n\rangle &= a^\dagger a |n\rangle - a a^\dagger |n\rangle \\ &= a^\dagger \sqrt{n} |n-1\rangle - a \sqrt{n+1} |n+1\rangle \\ &= n |n\rangle - (n+1) |n\rangle \\ &= (-1) |n\rangle \end{aligned}$$

This implies $[a, a^\dagger] = 1$. The action of these operators is equivalent to the corresponding operators for the harmonic oscillator. The general commutation relations are

$$[a_l, a_m^\dagger] = \delta_{lm} \quad (2.13)$$

$$[a_l, a_m] = [a_l^\dagger, a_m^\dagger] = 0 \quad (2.14)$$

The physical reason for this close analogy to the harmonic oscillator is that the energy levels of free particles in one mode have the same structure: the creation

¹Actually an additional particle is needed to make energy and momentum conservation possible. However, here we are only interested in the underlying principle.

of n particles each with energy E_0 corresponds to a total energy nE_0 . This corresponds to an n -fold excitation of a harmonic oscillator of energy $E_0 = \hbar\omega$

Obviously one finds for Bosons

$$\begin{aligned} a_l^\dagger \mathcal{H}^{(N)} &\longrightarrow \mathcal{H}^{(N+1)} \\ a_l \mathcal{H}^{(N)} &\longrightarrow \mathcal{H}^{(N-1)} \\ a_l |0\rangle &= 0 = \text{Number!} \end{aligned}$$

Here the first two equations define a mapping from Fock space into Fock space. The last equation defines the vacuum state and essentially states that one cannot annihilate a particle if there is none.

Because of Pauli's principle it is not possible to have two Fermions in the same state. Therefore, the Fermion creation operators need to fulfill $(a_l^\dagger)^2 = 0$. This can be achieved through the anticommutation relations

$$\{a_l^\dagger, a_m^\dagger\} = \{a_l, a_m\} = 0 \quad \{a_l, a_m^\dagger\} = \delta_{lm} \quad (2.15)$$

where the anticommutator is defined by $\{A, B\} := AB + BA$.

2.1.3 The Field Operator

How can one calculate expectation values using creation operators? Let us first consider an ordinary 1-particle operator $\hat{O}(\hat{x}, \hat{p})$. In the theory of a single particle one has

$$\langle \hat{O} \rangle = \int \psi^*(\hat{x}) \hat{O} \psi(x) d^3x$$

If we have N particles in this state the expectation value of \hat{O} is of course given by

$$\langle \hat{O} \rangle = \sum_{i=1}^N \langle \hat{O} \rangle_{1\text{-particle}}$$

This suggests to define the expectation value of a 1-particle operator in the framework of many-particle theory as follows,

$$\begin{aligned} \langle \hat{O}_N \rangle &:= \sum_{i=1}^N \langle \hat{O}(\hat{x}_i, \hat{p}_i) \rangle \\ &= \sum_{i=1}^N \int d^3x_1 \dots d^3x_n \psi_N^*(x_1, \dots, x_N) \hat{O}_N \psi(x_1, \dots, x_n) \quad (2.16) \end{aligned}$$

Consider for instance the energy and the corresponding equations for the eigenvalues,

$$H(\hat{x}, \hat{p}) \psi_a(x) = E_a \psi_a(x) \quad (2.17)$$

For a n -particle state $|n_a, \dots, n_{a'}, \dots, n_{a''}\rangle$ the energy is given by

$$\begin{aligned} E &= E_a n_a + \dots + E_{a'} n_{a'} + \dots \\ &= \int d^3x \psi_a^*(x) H(\hat{x}, \hat{p}) \psi_a(x) n_a + \int d^3x \psi_{a'}^*(x) H(\hat{x}, \hat{p}) \psi_{a'}(x) n_{a'} + \dots \end{aligned} \quad (2.18)$$

The particle numbers $n_a, n_{a'}, \dots$ can be calculated with the aid of creation/annihilation operators. To do so we introduce the operator

$$\boxed{\hat{N}_a = a_a^\dagger a_a} \quad (2.19)$$

This is the Number operator for particles in state a . Its physical meaning is revealed by the following calculation.

$$\begin{aligned} \langle \dots, n_a, \dots | \hat{N}_a | \dots, n_a, \dots \rangle &= \langle \dots, n_a, \dots | a_a^\dagger a_a | \dots, n_a, \dots \rangle \\ &= \langle \dots, n_a, \dots | a_a^\dagger \sqrt{n_a} | \dots, (n_a - 1), \dots \rangle \\ &= \langle \dots, n_a, \dots | \sqrt{(n_a - 1) + 1} \sqrt{n_a} | \dots, n_a, \dots \rangle = n_a \langle \dots | \dots \rangle = n_a \end{aligned} \quad (2.20)$$

And therefore we have for E :

$$\begin{aligned} E &= \int d^3x \psi_a^*(x) H(\hat{x}, \hat{p}) \psi_a(x) \langle \dots, n_a, \dots, n_{a'} | a_a^\dagger a_a | \dots, n_a, \dots, n_{a'} \rangle \\ &+ \int d^3x \psi_{a'}^*(x) H(\hat{x}, \hat{p}) \psi_{a'}(x) \langle \dots, n_a, \dots, n_{a'} | a_{a'}^\dagger a_{a'} | \dots, n_a, \dots, n_{a'} \rangle \\ &+ \dots \end{aligned} \quad (2.21)$$

Important:

In this expression the operators a_a, a_a^\dagger do only act on the many particle states, not on the wavefunction $\psi_a(x)$.

Contrarywise, $H(\hat{x}, \hat{p})$ acts only on $\psi_a(x)$ but not on $|\dots, n_a, \dots\rangle$.

The number representation of many-particle states can be considered as a kind of bookkeeping procedure to simplify the combinatorics associated with the symmetrization/antisymmetrization of many-particle states.

We therefore can rewrite the expression for E as

$$E = \langle \dots, n_a, n_{a'}, \dots | \int d^3x \psi_a^*(x) H(\hat{x}, \hat{p}) \psi_a(x) a_a^\dagger a_a | \dots, n_a, n_{a'}, \dots \rangle + \dots \quad (2.22)$$

The *field operator* is defined by $\Psi(x) := \sum_a a_a \psi_a(x)$ and allows us to rewrite the mean energy as

$$E = \langle \dots, n_a, n_{a'}, \dots | \int d^3x \Psi^\dagger(x) H(\hat{x}, \hat{y}) \Psi(x) | \dots, n_a, n_{a'}, \dots \rangle \quad (2.23)$$

Proof:

$$\begin{aligned} \langle \dots | \int d^3x \Psi^\dagger H \Psi | \dots \rangle &= \langle \dots | \int d^3x \sum_a \psi_a^*(x) a_a^\dagger H \sum_b \psi_b(x) a_b | \dots \rangle \quad (2.24) \\ &= \sum_{a,b} \underbrace{\int d^3x \psi_a^* H \psi_b}_{= E_b \int \psi_a^* \psi_b = E_b \delta_{ab}} \langle \dots | a_a^\dagger a_b | \dots \rangle = \sum_a E_a \langle \dots | a_a^\dagger a_a | \dots \rangle = \sum_a E_a n_a = E \end{aligned}$$

One generally can express the expectation value of any 1-particle operator $\hat{O}(\hat{x}, \hat{p})$ in a Fock state $|\psi\rangle$ by

$$\langle \hat{O}(\hat{x}, \hat{p}) \rangle = \langle \psi | \int d^3x \Psi^\dagger(x) \hat{O}(\hat{x}, \hat{p}) \Psi(x) | \psi \rangle \quad (2.25)$$

This holds for any state in Fock space, e.g.,

$$|\psi\rangle = |1_a\rangle + |2_{a'}\rangle \rightarrow \langle \psi | \Psi^\dagger \Psi | \psi \rangle = \langle 1_a | \Psi^\dagger \Psi | 1_a \rangle + \langle 2_{a'} | \Psi^\dagger \Psi | 2_{a'} \rangle \quad (2.26)$$

Remarks on the field operator:

- In a sense, $\Psi(x)$ is both an operator and a 1-particle state:

$$\Psi(x) = \sum_a a_a \psi_a(x) \quad (2.27)$$

a_a : Operator in Fock space

$\psi_a(x)$: 1-particle state

One could (and sometimes does) write $\psi_a(x)$ as $\langle x | \psi_a \rangle_{1\text{-particle}}$

$$\rightarrow \Psi(x) = \langle x | \sum_a a_a | \psi_a \rangle_{\text{Eint.}} = \langle x | \Psi \rangle_{\text{Eint.}} \quad (2.28)$$

- Interpretation of $\Psi(x)$: What is the meaning of the state $\Psi^\dagger(x) |0\rangle$? In the following we will see that it is a very special 1-particle state. To see this we

first consider a perfectly localized state in $\mathcal{H}^{(1)}$ (actually, perfectly localized states are not normalizable, but this doesn't matter for us):

$$\begin{aligned}\phi_{x_0}(x) &= \delta(x - x_0) = \sum_n c_n \phi_n(x), \quad \{\phi_n\} = \text{basis of } \mathcal{H}^{(1)} \\ c_n &= \int d^3x \phi_n^*(x) \phi_{x_0}(x) = \int d^3x \phi_n^*(x) \delta(x - x_0) = \phi_n^*(x_0) \\ \rightarrow \phi_{x_0}(x) &= \sum_n \phi_n^*(x_0) \phi_n(x) \quad (\text{completeness relation})\end{aligned}\tag{2.29}$$

The Fock-space representation of this state is given by

$$\sum_n \phi_n^*(x_0) |1_n\rangle = \sum_n \phi_n^*(x_0) a_n^\dagger |0\rangle = \Psi^\dagger(x_0) |0_n\rangle\tag{2.30}$$

Thus, $\Psi^\dagger(x_0)$ creates a particle at the position x_0 .

$\Psi(x_0)$ annihilates a particle at the position x_0 .

We therefore can (formally) introduce a state $|1_x\rangle \equiv \Psi^\dagger(x)|0\rangle$ which corresponds to one particle at position x . This allows us to find the mode function of a given state in much the same way as in ordinary quantum mechanics when the state is given by a Dirac ket $|\phi\rangle$. The corresponding wavefunction is then given by $\langle x|\phi\rangle$. The same state can be represented in Fock space as “one particle in mode ϕ ”, i.e., $|1_\phi\rangle$. To calculate the corresponding wavefunction one simply forms $\langle 1_x|1_\phi\rangle = \langle 0|\Psi(x)|1_\phi\rangle$. In general, $|1_x\rangle$ projects onto the one-particle wavefunction of a given state $|\psi\rangle$ in Fock space. A two-particle wavefunction can be calculated like $\langle 1_x 1_y|\psi\rangle$ and so on.

- Commutation relations for Bosons:

$$\begin{aligned}[\Psi(x), \Psi^\dagger(x)] &= \left[\sum_l a_l \psi_l(x), \sum_n a_n^\dagger \psi_n^*(y) \right] = \sum_{l,n} \psi_l(x) \psi_n^*(y) \underbrace{[a_l, a_m^\dagger]}_{\delta_{ln}} \\ &= \sum_n \psi_n(x) \psi_n^*(y) = \delta(x - y) \quad (\text{for Bosons})\end{aligned}\tag{2.31}$$

$$\boxed{[\Psi(x), \Psi^\dagger(y)] = \delta(x - y)}\tag{2.32}$$

$$\boxed{[\Psi^\dagger(x), \Psi^\dagger(y)] = [\Psi(x), \Psi(y)] = 0}\tag{2.33}$$

For Fermions we have analogously

$$\begin{aligned}\{a_l, a_m^\dagger\} &= \delta_{lm} = a_l a_m^\dagger + a_m a_l^\dagger \\ \rightarrow \boxed{\{\Psi(x), \Psi^\dagger(y)\} &= \delta(x - y)}\end{aligned}\tag{2.34}$$

$$\boxed{\{\Psi^\dagger(x), \Psi^\dagger(y)\} = \{\Psi(x), \Psi(y)\} = 0}\tag{2.35}$$

2.1.4 Second Quantization

The construction of field operators described above is mathematically equivalent to a second quantization procedure applied on the Schrödinger wavefunction.

Canonical quantization of a classical particle:

Using the Lagrange function $L(x, \dot{x})$ one can define the canonical momentum by

$$p = \frac{\partial L}{\partial \dot{x}} \quad (2.36)$$

Quantization is achieved by re-interpreting x and p as operators and demanding the commutation relations

$$[\hat{x}, \hat{p}] = i\hbar \quad (2.37)$$

The same procedure can be done for a field:

For a Schrödinger particle one can introduce the Lagrange density

$$\mathcal{L}(\psi, \psi^*, \dot{\psi}, \dot{\psi}^*) = i\hbar\psi^*\dot{\psi} - \frac{\hbar^2}{2m}\nabla\psi^*\nabla\psi - \psi^*V\psi \quad (2.38)$$

The Euler-Lagrange equation for this Lagrangean leads to the complex conjugate of the Schrödinger equation,

$$\partial_t \frac{\partial \mathcal{L}}{\partial \dot{\psi}} = -\partial_i \frac{\partial \mathcal{L}}{\partial (\partial_i \psi)} + \frac{\partial \mathcal{L}}{\partial \psi} \quad (2.39)$$

$$\rightarrow \partial_t i\hbar\psi^* = \partial_i^2 \frac{\hbar^2}{2m}\psi^* - V\psi^* \quad (2.40)$$

One can define the canonically conjugate momentum density by

$$\pi(x) = \frac{\partial \mathcal{L}}{\partial \dot{\psi}(x)} = i\hbar\psi^*(x) \quad (2.41)$$

If one applies again the canonical quantization scheme one finds

$$\begin{aligned} \psi(x), \pi(x) &\rightarrow \Psi(x), \Pi(x) \\ [\Psi(x), \Pi(y)] &= i\hbar\delta(x-y) \rightarrow [\Psi(x), \Psi^\dagger(y)] = \delta(x-y) \end{aligned} \quad (2.42)$$

This means that, by applying the canonical quantization procedure a second time, we end up with the same commutation relations as that derived from the construction of the field operator from creation/annihilation operators.

The method of second quantization is most popular method used to construct a many-particle theory of fields. However, it has the disadvantage that the construction of Fock space and the meaning of the field operator is not as obvious as in the previous procedure.

2.1.5 Summary of the construction of a many-particle theory

Recipe to construct a many-particle theory (quantum field theory) using the example of the Schrödinger field:

- (i) Choose stationary solutions $\varphi_n(x)$ of a 1-particle theory with $\langle \varphi_n | \varphi_m \rangle = \delta_{nm}$. The scalar product should be time-independent since otherwise the meaning of the modes, and therefore of the creation and annihilation operators, would be time-dependent. The interpretation of operators in Schrödinger picture would then be more difficult.

$$\begin{aligned} i\hbar \partial_t \psi &= H\psi, \quad \langle \psi | \psi' \rangle = \int d^3x \psi^* \psi' \\ \partial_t \langle \psi | \psi' \rangle &= \int d^3x \left(\dot{\psi}^* \psi' + \psi^* \dot{\psi}' \right) = \int d^3x \left\{ \frac{i}{\hbar} (H\psi^*) \psi' + \psi^* \frac{-i}{\hbar} H\psi' \right\} \\ &= \frac{i}{\hbar} \int d^3x \{ \psi^* (H\psi') - \psi^* (H\psi') \} = 0 \end{aligned} \quad (2.43)$$

where we have used the hermitean conjugate Schrödinger equation and the hermiticity of H with respect to the scalar product.

- (ii) Replace the general solution of the 1-particle equations of motion,

$$\psi(x) = \sum_n c_n \varphi_n(x), \quad (2.44)$$

by the field operator

$$\Psi(x) = \sum_n a_n \varphi_n(x) \quad \text{with} \quad [a_n, a_m^\dagger] = \delta_{nm} \quad (2.45)$$

($\{a_n, a_m^\dagger\} = \delta_{nm}$ for Fermions, respectively).
 $\Psi^\dagger(x)$ creates a particle at position x .

2.1.6 Quantization in Coulomb Gauge

The Lagrangean density \mathcal{L} of the electromagnetic field is given by

$$\mathcal{L} \propto \mathbf{E}^2 - c^2 \mathbf{B}^2 = (-\nabla\phi - \dot{\mathbf{A}})^2 - c^2 (\text{curl } \mathbf{A})^2 \quad (2.46)$$

\mathcal{L} does not depend on derivatives of the fields \mathbf{E} and \mathbf{B} . It therefore does only lead to Maxwell's equations if it is considered as a function of the potentials. The field equations in Coulomb gauge ($\text{div } \mathbf{A} = 0$) are

$$\square \mathbf{A} = \mu_0 \mathbf{j} - \frac{1}{c^2} \nabla \dot{\phi}, \quad \Delta \phi = -\frac{\rho}{\epsilon_0} \quad (2.47)$$

The equation for ϕ is not dynamical!

In vacuum the corresponding equations read

$$\square \mathbf{A} = -\frac{1}{c^2} \nabla \dot{\phi}, \quad \Delta \phi = 0 \quad (2.48)$$

Boundary conditions: $\phi \xrightarrow{r \rightarrow \infty} 0 \implies \phi = 0 \implies \square \mathbf{A} = 0$

Solution:

$$\mathbf{A}(\mathbf{x}, t) = \mathbf{A}^{(+)}(\mathbf{x}, t) + \mathbf{A}^{(-)}(\mathbf{x}, t) \quad (2.49)$$

$$= \underbrace{\int d^3k \sum_{\sigma=1,2} A_{k\sigma} \overbrace{\epsilon_{\sigma k} N_{\sigma k} \exp(-i\omega_k t) \exp(i\mathbf{k}\mathbf{x})}^{\text{mode function}}}_{\mathbf{A}^{(+)}(\mathbf{x}, t)} + \underbrace{\text{c.c.}}_{\mathbf{A}^{(-)}(\mathbf{x}, t)} \quad (2.50)$$

Here we have introduced

- the expansion coefficients $A_{k\sigma}$
- the polarization vectors $\epsilon_{\sigma k}$
- the normalization coefficients $N_{\sigma k}$
- $\mathbf{A}^{(+)}(\mathbf{x}, t)$ denotes the so-called „positive frequency part“ ($\propto \exp(-i\omega_k t)$) which only contains annihilation operators.

We know that $e^{i\mathbf{k}\mathbf{x}}$ is a complete basis (because of Fourier transformation, and leaving aside issues of normalization). Since all possible factors of $e^{i\mathbf{k}\mathbf{x}}$ already appear in $\mathbf{A}^{(+)}(\mathbf{x}, t)$, it is sufficient to consider only $\mathbf{A}^{(+)}(\mathbf{x}, t)$. Since the classical field $\mathbf{A}(\mathbf{x}, t)$ is real, the corresponding quantum field operator $\hat{\mathbf{A}}$ must be hermitean. Because of $[a, a^\dagger] = 1$ it is clear that a cannot be hermitean. Thus, the full operator $\hat{\mathbf{A}}$ cannot be composed out of annihilation operators alone. This is another (and more important) reason to use $\mathbf{A}^{(+)}(\mathbf{x}, t)$ instead of $\mathbf{A}(\mathbf{x}, t)$ for the quantization procedure.

Let now $\mathbf{A}^{(+)}(\mathbf{x}, t)$ and $\mathbf{A}'^{(+)}(\mathbf{x}, t)$ be solutions of $\square \mathbf{A} = 0$. A conserved scalar product is then given by

$$\left\langle \mathbf{A}^{(+)}(\mathbf{x}, t) \left| \mathbf{A}'^{(+)}(\mathbf{x}, t) \right. \right\rangle = -i \frac{\epsilon_0}{\hbar} \int d^3x \left\{ \underbrace{\dot{\mathbf{A}}^{(+)*}}_{\dot{\mathbf{A}}^{(-)}} \mathbf{A}'^{(+)} - \underbrace{\mathbf{A}^{(+)*}}_{\mathbf{A}^{(-)}} \dot{\mathbf{A}}'^{(+)} \right\}$$

The prefactor appears out of dimensional considerations:

$$[\mathbf{A}] = \frac{Js}{Cm} \implies [\langle \mathbf{A} | \mathbf{A}' \rangle] = 1$$

Proof of time-independence of the scalar product:

$$\begin{aligned} \partial_t \langle \mathbf{A} | \mathbf{A} \rangle &= -i \frac{\epsilon_0}{\hbar} \int d^3x \left\{ \ddot{\mathbf{A}}^* \mathbf{A}' + \dot{\mathbf{A}}^* \dot{\mathbf{A}}' - \dot{\mathbf{A}}^* \dot{\mathbf{A}}' - \mathbf{A}^* \ddot{\mathbf{A}}' \right\} \stackrel{!}{=} 0 \\ &\quad \text{use } \frac{1}{c^2} \ddot{\mathbf{A}} - \Delta \mathbf{A} = 0 \\ \implies \partial_t \langle \mathbf{A} | \mathbf{A} \rangle &= -i \frac{\epsilon_0}{\hbar} c^2 \int d^3x \{ (\Delta \mathbf{A}^*) \mathbf{A}' - \mathbf{A}^* (\Delta \mathbf{A}') \} \\ &= -i \frac{\epsilon_0}{\hbar} c^2 \int d^3x \{ \mathbf{A}^* (\Delta \mathbf{A}') - (\Delta \mathbf{A}^*) \mathbf{A}' \} \\ &= 0 \end{aligned}$$

where we used $\int (\nabla_i \nabla_i \mathbf{A}_j^*) \mathbf{A}'_j = - \int \nabla_i \mathbf{A}_j^* \nabla_i \mathbf{A}'_j = \int \mathbf{A}_j^* \nabla_i \nabla_i \mathbf{A}'_j$

Normalization of plane waves $N_{k\sigma} \boldsymbol{\epsilon}_{k\sigma} \exp(i\mathbf{k}\mathbf{x} - i\omega_k t) = \varphi_{k\sigma}(\mathbf{x}, t)$

$$\begin{aligned} \langle \varphi_{k\sigma} | \varphi_{k'\sigma'} \rangle &\stackrel{!}{=} \delta_{\sigma\sigma'} \delta(\mathbf{k} - \mathbf{k}') \\ &= -i \frac{\epsilon_0}{\hbar} \int d^3x N_{k\sigma} N_{k'\sigma'} \boldsymbol{\epsilon}_{k\sigma}^* \boldsymbol{\epsilon}_{k'\sigma'} \\ &\quad \left\{ e^{-i\mathbf{k}\mathbf{x}} e^{i\omega_k t} (i\omega_k) e^{i\mathbf{k}'\mathbf{x}} e^{-i\omega_{k'} t} \right. \\ &\quad \left. - e^{-i\mathbf{k}\mathbf{x}} e^{i\omega_k t} (-i\omega_{k'}) e^{i\mathbf{k}'\mathbf{x}} e^{-i\omega_{k'} t} \right\} \end{aligned}$$

with $\int d^3x e^{i(\mathbf{k}-\mathbf{k}')\mathbf{x}} = (2\pi)^3 \delta(\mathbf{k} - \mathbf{k}')$ it follows that

$$\begin{aligned} \langle \varphi_{k\sigma} | \varphi_{k'\sigma'} \rangle &= \frac{\epsilon_0}{\hbar} (2\pi)^3 \underbrace{\boldsymbol{\epsilon}_{k\sigma}^* \boldsymbol{\epsilon}_{k\sigma'}}_{\delta_{\sigma,\sigma'}} \delta(\mathbf{k} - \mathbf{k}') N_{k\sigma} N_{k\sigma'} 2\omega_k \\ &= \frac{\epsilon_0}{\hbar} (2\pi)^3 2\omega_k N_{k\sigma}^2 \delta_{\sigma,\sigma'} \delta(\mathbf{k} - \mathbf{k}') \\ &\stackrel{!}{=} \delta_{\sigma,\sigma'} \delta(\mathbf{k} - \mathbf{k}') \end{aligned}$$

$$\implies N_{k\sigma} = \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_k}}$$

for $t = 0$ it follows that ² for $\mathbf{A}(\mathbf{x})$:

$$\mathbf{A}(\mathbf{x}) = \int d^3k \sum_{\sigma} A_{k\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_k}} \boldsymbol{\epsilon}_{k\sigma} \exp(i\mathbf{k}\mathbf{x}) + \text{c.c.} \quad (2.51)$$

Quantization: replace the amplitude $A_{k\sigma}$ by the annihilation operator $a_{k\sigma}$ with

$$[a_{k\sigma}, a_{k'\sigma'}^{\dagger}] = \delta_{\sigma,\sigma'} \delta(\mathbf{k} - \mathbf{k}')$$

$a_{k\sigma}^{\dagger}$ creates a photon (= quantum of light) with momentum $\hbar\mathbf{k}$ and polarization vector $\boldsymbol{\epsilon}_{k\sigma}$.

In contrast to the classical theory of electromagnetism, intensity and energy of a light beam appear in quantized portions.

Photons are Bosons and have spin 1, but they have only **two** possible values for the magnetic quantum number, $m = +1, -1$. These values correspond to left- and right-circularly polarized plane waves. $m = 0$ (\Rightarrow longitudinally polarized photons $\Leftrightarrow \boldsymbol{\epsilon}_{k\sigma} \parallel \mathbf{k}$) is not possible because of the vanishing mass of photons (see, e.g., Sec. 14.5 of Ref. [9]).

The operators for the electric and magnetic field can easily be derived from that for the vector potential:

$$\hat{\mathbf{E}}(\mathbf{x}) \Big|_{t=0} = -\dot{\hat{\mathbf{A}}}(\mathbf{x}) \Big|_{t=0} = i \int d^3k \sum_{\sigma} a_{k\sigma} \sqrt{\frac{\hbar\omega_k}{2(2\pi)^3 \epsilon_0}} \boldsymbol{\epsilon}_{k\sigma} e^{i\mathbf{k}\mathbf{x}} + \text{H.c.} \quad (2.52)$$

$$\hat{\mathbf{B}}(\mathbf{x}) = \text{rot} \hat{\mathbf{A}}(\mathbf{x}) = i \int d^3k \sum_{\sigma} a_{k\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_k}} (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma}) e^{i\mathbf{k}\mathbf{x}} + \text{H.c.} \quad (2.53)$$

The Hamiltonian can be found using the classical expression for the energy density:

$$\begin{aligned} \hat{H} &= \frac{\epsilon_0}{2} \int d^3x \left(\hat{\mathbf{E}}^2(\mathbf{x}) + c^2 \hat{\mathbf{B}}^2(\mathbf{x}) \right) = \dots \\ &= \int d^3k \sum_{\sigma} \frac{\hbar\omega_k}{2} \left\{ a_{k\sigma}^{\dagger} a_{k\sigma} + a_{k\sigma} a_{k\sigma}^{\dagger} \right\} \\ &= \int d^3k \sum_{\sigma} \hbar\omega_k \left\{ a_{k\sigma}^{\dagger} a_{k\sigma} + \frac{1}{2} \right\} \end{aligned} \quad (2.54)$$

²We set $t = 0$ to derive the time-independent vector potential operator in Schrödinger picture. One can also consider $t \neq 0$ to derive the corresponding operator in interaction picture.

(\Leftrightarrow sum over harmonic oscillators)

The Heisenberg equations of motion for operators (in Heisenberg picture) are generally given by

$$i\hbar\partial_t\hat{O}(t) = [\hat{O}(t), \hat{H}] \quad (2.55)$$

so that we can deduce

$$\begin{aligned} i\hbar\dot{E}_i(\mathbf{x}, t) &= \left[E_i(\mathbf{x}, t), \int d^3k \sum_{\sigma} \hbar\omega_k \left\{ a_{k\sigma}^{\dagger} a_{k\sigma} + \frac{1}{2} \right\} \right] \\ &= i \int d^3k' \sum_{\sigma'} \sqrt{\frac{\hbar\omega_{k'}}{2(2\pi)^3\epsilon_0}} \\ &\quad \left[a_{k'\sigma'} e^{i\mathbf{k}'\mathbf{x}} (\boldsymbol{\epsilon}_{k'\sigma'})_i - a_{k'\sigma'}^{\dagger} e^{-i\mathbf{k}'\mathbf{x}} (\boldsymbol{\epsilon}_{k'\sigma'}^*)_i, \int d^3k \sum_{\sigma} \hbar\omega_k \left\{ a_{k\sigma}^{\dagger} a_{k\sigma} + \frac{1}{2} \right\} \right] \\ &= i \int d^3k' \sum_{\sigma'} \int d^3k \sum_{\sigma} \sqrt{\frac{\hbar\omega_{k'}}{2(2\pi)^3\epsilon_0}} \hbar\omega_k \\ &\quad \left\{ e^{i\mathbf{k}'\mathbf{x}} (\boldsymbol{\epsilon}_{k'\sigma'})_i \delta(\mathbf{k} - \mathbf{k}') \delta_{\sigma\sigma'} a_{k\sigma} - e^{-i\mathbf{k}'\mathbf{x}} (\boldsymbol{\epsilon}_{k\sigma}^*)_i a_{k'\sigma'}^{\dagger} (-\delta_{\sigma\sigma'} \delta(\mathbf{k} - \mathbf{k}')) \right\} \\ &\Rightarrow \dot{\mathbf{E}} = \int d^3k \sum_{\sigma} \omega_k^2 \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}} \left(a_{k\sigma} e^{i\mathbf{k}\mathbf{x}} \boldsymbol{\epsilon}_{k\sigma} + a_{k\sigma}^{\dagger} e^{-i\mathbf{k}\mathbf{x}} \boldsymbol{\epsilon}_{k\sigma}^* \right) \quad (2.56) \end{aligned}$$

Let us now calculate $(\text{curl } \mathbf{B})_i$:

$$\begin{aligned} (\text{curl } \mathbf{B})_i &= i \int d^3k \sum_{\sigma} \varepsilon_{ijl} \frac{\partial}{\partial x_j} a_{k\sigma} (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma})_l e^{i\mathbf{k}\mathbf{x}} \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}} \\ &\quad - i \int d^3k \sum_{\sigma} \varepsilon_{ijl} \frac{\partial}{\partial x_j} a_{k\sigma}^{\dagger} (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma}^*)_l e^{-i\mathbf{k}\mathbf{x}} \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}} \\ \text{curl } \mathbf{B} &= - \int d^3k \sum_{\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}} \left(\mathbf{k} \times (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma}) a_{k\sigma} e^{i\mathbf{k}\mathbf{x}} + \mathbf{k} \times (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma}^*) a_{k\sigma}^{\dagger} e^{-i\mathbf{k}\mathbf{x}} \right) \quad (2.57) \end{aligned}$$

With $\mathbf{k} \cdot \boldsymbol{\epsilon}_{k\sigma} = 0$ we also find

$$\mathbf{k} \times (\mathbf{k} \times \boldsymbol{\epsilon}_{k\sigma}) = \mathbf{k}(\mathbf{k} \cdot \boldsymbol{\epsilon}_{k\sigma}) - \boldsymbol{\epsilon}_{k\sigma} k^2 = -\boldsymbol{\epsilon}_{k\sigma} k^2 \quad (2.58)$$

so that

$$\text{curl } \mathbf{B} = \int d^3k \sum_{\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3 \varepsilon_0 \omega_k}} k^2 \{a_{k\sigma} e^{i\mathbf{k}\mathbf{x}} \boldsymbol{\varepsilon}_{k\sigma} + a_{k\sigma}^{\dagger} e^{-i\mathbf{k}\mathbf{x}} \boldsymbol{\varepsilon}_{k\sigma}^*\} \quad (2.59)$$

$$\Rightarrow \text{curl } \mathbf{B} = \frac{1}{c^2} \dot{\mathbf{E}} = \mu_0 \varepsilon_0 \dot{\mathbf{E}} \quad (2.60)$$

This is one of the inhomogeneous Maxwell equations for $\mathbf{j} = 0$ (otherwise we have $\text{curl } \mathbf{B} = \frac{1}{c^2} \dot{\mathbf{E}} = \mu_0 \{\mathbf{j} + \varepsilon_0 \dot{\mathbf{E}}\}$). Analogously one can show that

$$\begin{aligned} i\hbar \dot{\mathbf{B}} &= [\mathbf{B}, H] = \dots \\ \Rightarrow \dot{\mathbf{B}} &= -\text{curl } \mathbf{E} \end{aligned} \quad (2.61)$$

Because $\mathbf{k} \cdot \boldsymbol{\varepsilon}_{k\sigma} = \mathbf{k} \cdot (\mathbf{k} \times \boldsymbol{\varepsilon}_{k\sigma}) = 0$

$$\text{div } \mathbf{B} = 0 \quad (2.62)$$

and

$$\text{div } \mathbf{E} = 0 \quad \left(= \frac{\rho}{\varepsilon_0} \right) \quad (2.63)$$

\Rightarrow *The field operators fulfill Maxwell's equations in a vacuum*

The equations of motions are easily solved if one considers the individual modes,

$$\begin{aligned} i\hbar \dot{a}_{k\sigma} &= [a_{k\sigma}, H] \\ &= \left[a_{k\sigma}, \int d^3k' \sum_{\sigma'} \hbar \omega_{k'} \left(a_{k'\sigma'}^{\dagger} a_{k'\sigma'} + \frac{1}{2} \right) \right] \\ &= \int d^3k' \sum_{\sigma'} \hbar \omega_{k'} \delta_{\sigma\sigma'} \delta(\mathbf{k} - \mathbf{k}') a_{k'\sigma'} \\ &= \hbar \omega_k a_{k\sigma} \end{aligned} \quad (2.64)$$

The solution is

$$a_{k\sigma} = e^{-i\omega_k t} a_{k\sigma}(0)$$

where $a_{k\sigma}(0)$ is the Schrödinger operator for the corresponding mode. The solution has the same structure as the respective classical wave.

Remark regarding the factor of $\frac{1}{2}$ in the Hamiltonian:

This term is constant and therefore does not have an effect on the field dynamics.

It represents the energy of the vacuum state and diverges mathematically. However, it depends on the boundary conditions imposed on the mode functions.

Example: Two mirrors at distance L (see Fig. 2.1). The (ideal) mirrors can be thought of as representing boundary conditions for the electromagnetic field. As a consequence, only discrete wavenumbers with $k_n = \frac{2\pi}{L}n$, $n \in \mathbb{N}$ instead of $|\mathbf{k}| \in \mathbb{R}$ are possible. Consequently, the sum over the modes in the Hamiltonian only runs over discrete wavenumbers and the vacuum energy is changed (though still divergent).

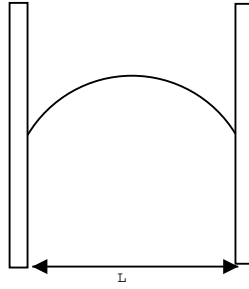


Figure 2.1: Sketch to the Casimir effect

If one subtracts the vacuum energy in free space from that between the two mirrors one finds a finite energy difference which depends on the distance L . This leads to an attractive force, the (*Casimir force*), between the mirrors. The Casimir effect is extensively discussed in Ref. [8].

2.1.7 Remarks on the Quantization Procedure for other Gauges

The fact that we have only quantized $\hat{\mathbf{A}}$ but not ϕ is connected to our choice of Coulomb gauge.

Reason: $\square \mathbf{A} = 0 \quad \Delta \phi = 0$

$\Rightarrow \phi$ is not dynamical, the corresponding conjugate momentum vanishes. This can be understood with the aid of the Lagrangean:

$$\begin{aligned}
 L &= \frac{1}{2} \int (\mathbf{E}^2 - c^2 \mathbf{B}^2) d^3x \\
 &= \frac{1}{2} \int [(-\nabla \phi - \dot{\mathbf{A}})^2 - c^2 (\text{rot} \mathbf{A})^2] d^3x \\
 &= \frac{1}{2} \int [(\nabla \phi)^2 + 2\phi \text{div} \dot{\mathbf{A}} + \dot{\mathbf{A}}^2 - c^2 (\text{rot} \mathbf{A})^2] d^3x \\
 &= \int \mathcal{L} d^3x
 \end{aligned}$$

Coulomb gauge:

$$\operatorname{div} \mathbf{A} = 0 \implies \frac{\partial \mathcal{L}}{\partial \dot{\phi}} = 0$$

Lorentz gauge:

$$\operatorname{div} \mathbf{A} + \frac{1}{c^2} \dot{\phi} = 0 \implies L = \frac{1}{2} \int [(\nabla \phi)^2 - 2\phi \ddot{\phi} \frac{1}{c^2} + \dot{\mathbf{A}}^2 - c^2(\operatorname{rot} \mathbf{A}^2)] d^3x$$

If one now considers the action $S = \int L dt$ one can integrate by part the term $\propto \ddot{\phi}$ and arrives at a new Lagrangean density $\mathcal{L}' \propto \dot{\phi}^2$. One therefore gets $\partial \mathcal{L}' / \partial \dot{\phi} \neq 0$ so that the scalar canonical momentum is nonzero.

\implies in Lorentz gauge we do not only have to circularly polarized photons but also a scalar photon ($\iff \phi$) and a longitudinally polarized one ($\iff \operatorname{div} \mathbf{A} \neq 0$). The two new photons are unphysical and have to be removed by imposing appropriate constraints: $(\operatorname{div} \hat{\mathbf{A}} + \dot{\hat{\phi}}/c^2) |\psi\rangle \stackrel{!}{=} 0$ for physical states $|\psi\rangle$.

A unified prescription to quantize constrained systems has been developed (among others) by Dirac, but it is rather sophisticated (see, for instance, chapter 1 of Ref. [10]).

Chapter 3

Mode theory and linear optical elements

The invention of the laser has provided us with a source of highly coherent light and laid the foundation for many outstanding experiments in quantum optics which otherwise would have been impossible to perform. From an idealized theoretical point of view, a laser prepares a many-photon state in which all photons are in the same spatial (and polarization) mode. Hence, the state can approximately be written as

$$|\psi\rangle = f(a_\phi^\dagger)|0\rangle \quad (3.1)$$

where a_ϕ^\dagger is the creation operator for a photon in the spatial mode $\phi(\mathbf{x})$. The notion of a mode has been introduced in Chapter 2 and will be studied in detail in this chapter. For many applications it is sufficient to think of a laser mode as a plane wave; the mode function then reduces to that given in Eq. (2.51), i.e.,

$$\phi_{\mathbf{k},\sigma}(\mathbf{x}, t) = \sqrt{\frac{\hbar}{2(2\pi)^3\epsilon_0\omega_k}} \boldsymbol{\epsilon}_{k\sigma} \exp(i\mathbf{k}\mathbf{x})e^{-i\omega_k t}. \quad (3.2)$$

However, this is not always sufficient since a real laser beam has a finite width which increases over time. A better description can be given using the paraxial approximation (Sec. 1.8) which allows to describe TEM laser modes. The proper description of photon modes will be the topic of the first part of this chapter. At the end of the chapter we will also consider the possibility to work with non-orthogonal modes. This includes the Hong-Ou-Mandel dip and Kerr nonlinear media.

Ignoring the shape of a photon mode for the moment, Eq. (3.1) still represents a tremendously simplified form of the real state of a laser. In general, a laser emits photons in different modes and does not produce a pure state but a mixed state (see Chapter 7).

However, Eq. (3.1) nevertheless describes many experiments remarkably well and is the basis of many theoretical investigations in quantum optics. As a consequence, one often needs only to consider one or two modes which reduces the study of a quantized light field to one or two harmonic oscillators. This simple model allows for a convenient description of many experiments which do only employ so-called *linear optical elements*: mirrors, beam splitters, phase shifter, polarization rotators, quarter-wave and half-wave plates, polarizing beam splitters, and, to some extent, so-called nonlinear crystals. The second part of the chapter will provide an introduction to this important field.

3.1 Mode expansion of the quantized radiation field

The general construction of a quantum field theory presented in Chapter 2 included the introduction of a field operator which is defined as a sum over stationary mode functions times their corresponding annihilation operator. For the electromagnetic field this field operator corresponded to the positive-frequency part $\mathbf{A}^{(+)}(\mathbf{x})$ of the operator for the vector potential.

However, this is not the most general expansion. A better way is to admit a general orthonormal set of mode functions $\mathbf{A}_n(\mathbf{x})$. One example for such a mode function might be a laser pulse at some fixed time. Clearly, such a state will propagate in time so that the meaning of what the corresponding creation operator actually creates will change over time. While this may appear odd in Schrödinger picture it is actually quite natural if we formulate the theory in the Heisenberg picture (or in the interaction picture if the electromagnetic field is coupled to atoms). Following the derivations in Sec. 2.1.6 the positive-frequency part of the vector potential operator in Heisenberg picture can be written as

$$\begin{aligned}\mathbf{A}^{(+)}(\mathbf{x}, t) &= \int d^3k \sum_{\sigma} a_{k\sigma} \sqrt{\frac{\hbar}{2(2\pi)^3 \epsilon_0 \omega_k}} \boldsymbol{\epsilon}_{k\sigma} e^{i\mathbf{k}\cdot\mathbf{x}} e^{-i\omega_k t} \\ &= \int d^3k \sum_{\sigma} a_{k\sigma} \boldsymbol{\phi}_{\mathbf{k},\sigma}(\mathbf{x}, t)\end{aligned}\tag{3.3}$$

As has been shown in Sec. 2.1.6 this operator is a solution to Maxwell's equations since the mode functions are solutions and the annihilation operators are constant. As in any quantum field theory, the field operator can also be expanded in another set of mode functions,

$$\mathbf{A}^{(+)}(\mathbf{x}, t) = \sum_n \mathbf{A}_n(\mathbf{x}, t) a_n .\tag{3.4}$$

The classical fields $\mathbf{A}_n(\mathbf{x}, t)$ are simply a set of solutions of Maxwell's equations in vacuum; plane waves are the most prominent example. Furthermore, the requirement that they are positive-frequency modes restricts these solutions to be of the general form

$$\mathbf{A}_n(\mathbf{x}, t) = \int d^3k \sum_{\sigma} A_n(\mathbf{k}, \sigma) \boldsymbol{\varepsilon}_{\mathbf{k}, \sigma} e^{i\mathbf{k} \cdot \mathbf{x}} e^{-i\omega_k t} \quad (3.5)$$

in terms of an expansion in plane waves. Note that this also guarantees that the field modes are transverse, i.e., $\nabla \cdot \mathbf{A}_n = 0$.

There is a useful formal way of dealing with positive-frequency fields. Taking the time derivative of Eq. (3.5) one finds

$$\begin{aligned} i\hbar \partial_t \mathbf{A}_n(\mathbf{x}, t) &= \int d^3k \hbar \omega_k \sum_{\sigma} A_n(\mathbf{k}, \sigma) \boldsymbol{\varepsilon}_{\mathbf{k}, \sigma} e^{i\mathbf{k} \cdot \mathbf{x}} e^{-i\omega_k t} \\ &\equiv \hbar \hat{\omega} \int d^3k \sum_{\sigma} A_n(\mathbf{k}, \sigma) \boldsymbol{\varepsilon}_{\mathbf{k}, \sigma} e^{i\mathbf{k} \cdot \mathbf{x}} e^{-i\omega_k t} \\ &= \hbar \hat{\omega} \mathbf{A}_n(\mathbf{x}, t) \end{aligned} \quad (3.6)$$

This is a first-order differential equation in time that be used instead of the (second-order) wave equation to describe the dynamics of the Maxwell field. In its derivation we have introduced the *frequency operator* $\hat{\omega}$ which simply multiplies any Fourier component \mathbf{k} with the corresponding frequency $\omega_k = c|\mathbf{k}|$. In other words, it is diagonal in momentum space. Another operator that is diagonal in momentum space is the operator $-i\nabla$. It is then easy to show that the frequency operator can formally be written as $\hat{\omega} = c\sqrt{-\Delta}$. Such a non-polynomial function of a differential operator is called a pseudo-differential operator. The frequency operator is Hermitean with respect to the usual Schrödinger scalar product. With its aid we can write any positive-frequency solution to the wave equation or to Eq. (3.6) in the elegant form

$$\mathbf{A}_n(\mathbf{x}, t) = e^{-i\hat{\omega}t} \mathbf{A}_n(\mathbf{x}, 0) . \quad (3.7)$$

The commutation relations of the annihilation operators a_n are determined by the following

Theorem: For two normalized transverse positive-frequency solutions of the Maxwell equations, represented by vector potentials \mathbf{A}_n and \mathbf{A}_m , the commutator of the respective operators is given by

$$\boxed{[a_n, a_m^\dagger] = \langle \mathbf{A}_n | \mathbf{A}_m \rangle} , \quad (3.8)$$

where the conserved scalar product between two classical solutions of Maxwell's equations is given by

$$\langle \mathbf{A}_n | \mathbf{A}_m \rangle = \frac{i\epsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot \dot{\mathbf{A}}_m - \dot{\mathbf{A}}_n^* \cdot \mathbf{A}_m \right) . \quad (3.9)$$

It is worthwhile to note that this commutator is time independent even if the modes are not stationary because of the time independence of the scalar product. We remark that the scalar product between two positive-frequency modes can be reduced to ¹

$$\begin{aligned}\langle \mathbf{A}_n | \mathbf{A}_m \rangle &= \frac{i\epsilon_0}{\hbar} \int d^3x (\mathbf{A}_n^* \cdot (-i)\hat{\omega} \mathbf{A}_m - i\hat{\omega} \mathbf{A}_n^* \cdot \mathbf{A}_m) \\ &= \frac{i\epsilon_0}{\hbar} \int d^3x (\mathbf{A}_n^* \cdot (-i)\hat{\omega} \mathbf{A}_m - i\mathbf{A}_n^* \cdot \hat{\omega} \mathbf{A}_m) \\ &= \frac{2\epsilon_0}{\hbar} \int d^3x \mathbf{A}_n^* \cdot \hat{\omega} \mathbf{A}_m\end{aligned}$$

Proof of theorem: The field operators for the electromagnetic field can generally be expanded in normalized mode functions as ²

$$\hat{\mathbf{A}} = \sum_n \mathbf{A}_n(\mathbf{x}, t) a_n + \mathbf{A}_n^*(\mathbf{x}, t) a_n^\dagger \quad (3.10)$$

$$\hat{\mathbf{E}} = \sum_n \mathbf{E}_n(\mathbf{x}, t) a_n + \mathbf{E}_n^*(\mathbf{x}, t) a_n^\dagger \quad (3.11)$$

$$\hat{\mathbf{B}} = \sum_n \mathbf{B}_n(\mathbf{x}, t) a_n + \mathbf{B}_n^*(\mathbf{x}, t) a_n^\dagger \quad (3.12)$$

with $\mathbf{E}_n = -\dot{\mathbf{A}}_n$ and $\mathbf{B}_n = \nabla \times \mathbf{A}_n$. The operator a_n can then be represented in the form

$$a_n = \langle \mathbf{A}_n | \hat{\mathbf{A}} \rangle. \quad (3.13)$$

This also shows that the annihilation operator is time independent since both the mode function and the field operator are solutions to Maxwell's equations, so that their scalar product is time independent. Eq. (3.13) can be shown as follows.

$$\begin{aligned}\langle \mathbf{A}_n | \hat{\mathbf{A}} \rangle &= \sum_m a_m \langle \mathbf{A}_n | \mathbf{A}_m \rangle + a_m^\dagger \langle \mathbf{A}_n | \mathbf{A}_m^* \rangle \\ &= \sum_m a_m \delta_{m,n} + a_m^\dagger \times 0\end{aligned} \quad (3.14)$$

¹For later use we also remark that it can also be written in the form

$$\langle \mathbf{A}_n | \mathbf{A}_m \rangle = \frac{2\epsilon_0}{\hbar} \int d^3x \mathbf{E}_n^* \cdot \hat{\omega}^{-1} \mathbf{E}_m$$

²In this chapter mode theory is presented for the free electromagnetic field using the electric field operator \mathbf{E} . However, after the Power-Zienau-Woolley transformation (see Chapter 5) it is usually more convenient to deal with the dielectric displacement \mathbf{D} instead. This poses no problem since for free fields these operators are related by $\mathbf{D} = \epsilon_0 \mathbf{E}$ so that $\mathbf{D} = -\dot{\mathbf{A}}/\epsilon_0$.

The scalar product $\langle \mathbf{A}_n | \mathbf{A}_m^* \rangle$ vanishes because the modes are positive-frequency modes, i.e., they can be represented as $\mathbf{A}_n(\mathbf{x}, t) = \exp(-i\hat{\omega}t)\mathbf{A}_n(\mathbf{x}, 0)$. It therefore follows that

$$\begin{aligned} \langle \mathbf{A}_n | \mathbf{A}_m^* \rangle &= \frac{i\epsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot \dot{\mathbf{A}}_m^* - \dot{\mathbf{A}}_n^* \cdot \mathbf{A}_m^* \right) \\ &= \frac{i\epsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot i\hat{\omega} \mathbf{A}_m^* - i\hat{\omega} \mathbf{A}_n^* \cdot \mathbf{A}_m^* \right) \\ &= 0. \end{aligned} \quad (3.15)$$

With Eq. (3.13) we find

$$\begin{aligned} [a_n, a_m^\dagger] &= [\langle \mathbf{A}_n | \hat{\mathbf{A}} \rangle, \langle \hat{\mathbf{A}} | \mathbf{A}_m \rangle] \\ &= \left(\frac{i\epsilon_0}{\hbar} \right)^2 \int d^3x d^3x' [(\mathbf{A}_n^*(\mathbf{x}) \dot{\hat{\mathbf{A}}}(\mathbf{x}) - \dot{\mathbf{A}}_n^*(\mathbf{x}) \hat{\mathbf{A}}(\mathbf{x})), (\hat{\mathbf{A}}(\mathbf{x}') \dot{\mathbf{A}}_m(\mathbf{x}') - \dot{\hat{\mathbf{A}}}(\mathbf{x}') \mathbf{A}_m(\mathbf{x}'))] \\ &= \left(\frac{i\epsilon_0}{\hbar} \right)^2 \int d^3x d^3x' [-(\mathbf{A}_n^*(\mathbf{x}) \hat{\mathbf{E}}(\mathbf{x}) - \dot{\mathbf{A}}_n^*(\mathbf{x}) \hat{\mathbf{A}}(\mathbf{x})), (\hat{\mathbf{A}}(\mathbf{x}') \dot{\mathbf{A}}_m(\mathbf{x}') + \hat{\mathbf{E}}(\mathbf{x}') \mathbf{A}_m(\mathbf{x}'))] \end{aligned} \quad (3.16)$$

We can now exploit that the equal-time commutators $[A, A]$ and $[E, E]$ vanish, and that the remaining commutator is given by

$$[\mathbf{A}_i(\mathbf{x}), \mathbf{E}_j(\mathbf{x}')] = \frac{\hbar}{i\epsilon_0} \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \quad (3.17)$$

with the transverse delta function

$$\delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') = \int \frac{d^3k}{(2\pi)^3} \left(\delta_{ij} - \frac{\mathbf{k}_i \mathbf{k}_j}{\mathbf{k}^2} \right) \exp(i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}')) \quad (3.18)$$

This results in

$$\begin{aligned} [a_n, a_m^\dagger] &= \frac{i\epsilon_0}{\hbar} \int d^3x d^3x' \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \times \\ &\quad \left(\mathbf{A}_{n,i}^*(\mathbf{x}) \dot{\mathbf{A}}_{m,j}(\mathbf{x}') - \dot{\mathbf{A}}_{n,i}^*(\mathbf{x}) \mathbf{A}_{m,j}(\mathbf{x}') \right) \end{aligned} \quad (3.19)$$

All mode functions appearing in this expression are transverse ($\nabla \cdot \mathbf{A}_n = 0$ etc.). Since for transverse fields the identity

$$\int d^3x' \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \mathbf{A}_{m,j}(\mathbf{x}') = \mathbf{A}_{m,i}(\mathbf{x}) \quad (3.20)$$

holds, this expression is identical to the scalar product between the modes and therefor concludes the proof of Eq. (3.8) \square

The result (3.8) allows for a convenient expansion of all quantum field operators in terms of mode operators. For instance, the Hamiltonian of the quantized radiation field reads

$$\begin{aligned}
H &= \frac{\epsilon_0}{2} \int d^3x : \left(\widehat{\mathbf{E}}^2(\mathbf{x}) + c^2 \widehat{\mathbf{B}}^2(\mathbf{x}) \right) : \\
&= \frac{\epsilon_0}{2} \int d^3x \sum_{n,m} : \left((-\dot{\mathbf{A}}_n a_n - \dot{\mathbf{A}}_n^* a_n^\dagger) \cdot (-\dot{\mathbf{A}}_m a_m - \dot{\mathbf{A}}_m^* a_m^\dagger) \right. \\
&\quad \left. + c^2 (\nabla \times \mathbf{A}_n a_n + \nabla \times \mathbf{A}_n^* a_n^\dagger) \cdot (\nabla \times \mathbf{A}_m a_m + \nabla \times \mathbf{A}_m^* a_m^\dagger) \right) : \\
&= \frac{\epsilon_0}{2} \sum_{n,m} \left\{ a_n a_m \int \left(\dot{\mathbf{A}}_n \cdot \dot{\mathbf{A}}_m + c^2 \nabla \times \mathbf{A}_n \cdot \nabla \times \mathbf{A}_m \right) \right. \\
&\quad \left. + a_n^\dagger a_m \int \left(\dot{\mathbf{A}}_n^* \cdot \dot{\mathbf{A}}_m + c^2 \nabla \times \mathbf{A}_n^* \cdot \nabla \times \mathbf{A}_m \right) \right. \\
&\quad \left. + a_m^\dagger a_n \int \left(\dot{\mathbf{A}}_n \cdot \dot{\mathbf{A}}_m^* + c^2 \nabla \times \mathbf{A}_n \cdot \nabla \times \mathbf{A}_m^* \right) \right. \\
&\quad \left. + a_n^\dagger a_m^\dagger \int \left(\dot{\mathbf{A}}_n^* \cdot \dot{\mathbf{A}}_m^* + c^2 \nabla \times \mathbf{A}_n^* \cdot \nabla \times \mathbf{A}_m^* \right) \right\} \quad (3.21)
\end{aligned}$$

Now,

$$\begin{aligned}
c^2 \int \nabla \times \mathbf{A} \cdot \nabla \times \mathbf{A}' &= c^2 \int \nabla \times \nabla \times \mathbf{A} \cdot \mathbf{A}' \\
&= c^2 \int (-\Delta \mathbf{A} + \nabla(\nabla \cdot \mathbf{A})) \cdot \mathbf{A}' \\
&= \int (\hat{\omega}^2 \mathbf{A}) \cdot \mathbf{A}' \\
&= \int \hat{\omega} \mathbf{A} \cdot \hat{\omega} \mathbf{A}' \quad (3.22)
\end{aligned}$$

where the second-to-last line follows from the definition of the frequency operator and the transversality of the field modes, $\nabla \cdot \mathbf{A} = 0$. The last line is a consequence of the Hermiticity of the frequency operator with respect to the Schrödinger scalar product. These expressions allow us to further reduce the Hamiltonian. The coefficient for $a_n a_m$ (and for $a_n^\dagger a_m^\dagger$) vanishes:

$$\begin{aligned}
\int \left(\dot{\mathbf{A}}_n \cdot \dot{\mathbf{A}}_m + c^2 \nabla \times \mathbf{A}_n \cdot \nabla \times \mathbf{A}_m \right) &= \int \left((-i\hat{\omega} \mathbf{A}_n) \cdot (-i\hat{\omega} \mathbf{A}_m) + c^2 \nabla \times \mathbf{A}_n \cdot \nabla \times \mathbf{A}_m \right) \\
&= \int \left(-\hat{\omega} \mathbf{A}_n \cdot \hat{\omega} \mathbf{A}_m + c^2 \nabla \times \mathbf{A}_n \cdot \nabla \times \mathbf{A}_m \right) \\
&= 0 \quad (3.23)
\end{aligned}$$

In a similar manner, we find for the other coefficients

$$\begin{aligned}
\int \left(\dot{\mathbf{A}}_n^* \cdot \dot{\mathbf{A}}_m + c^2 \nabla \times \mathbf{A}_n^* \cdot \nabla \times \mathbf{A}_m \right) &= \int \left(\dot{\mathbf{A}}_n^* \cdot \dot{\mathbf{A}}_m + \hat{\omega} \mathbf{A}_n^* \cdot \hat{\omega} \mathbf{A}_m \right) \\
&= \int \left(\dot{\mathbf{A}}_n^* \cdot \dot{\mathbf{A}}_m + i\hat{\omega} \mathbf{A}_n^* \cdot \dot{\mathbf{A}}_m \right) \\
&= \int \left(-i\dot{\mathbf{A}}_n^* \cdot \hat{\omega} \mathbf{A}_m + i\hat{\omega} \mathbf{A}_n^* \cdot \dot{\mathbf{A}}_m \right) \\
&= \int \left(-i\dot{\mathbf{A}}_n^* \cdot \hat{\omega} \mathbf{A}_m + i\mathbf{A}_n^* \cdot \hat{\omega} \dot{\mathbf{A}}_m \right) \\
&= \frac{\hbar}{\varepsilon_0} \langle \mathbf{A}_n | \hat{\omega} \mathbf{A}_m \rangle
\end{aligned} \tag{3.24}$$

so that

$$\begin{aligned}
H &= \frac{\hbar}{2} \sum_{n,m} \left(a_n^\dagger a_m \langle \mathbf{A}_n | \hat{\omega} \mathbf{A}_m \rangle + a_m^\dagger a_n \langle \mathbf{A}_m | \hat{\omega} \mathbf{A}_n \rangle \right) \\
&= \hbar \sum_{n,m} a_n^\dagger a_m \langle \mathbf{A}_n | \hat{\omega} \mathbf{A}_m \rangle
\end{aligned} \tag{3.25}$$

Often monochromatic modes are chosen so that $\hat{\omega} \mathbf{A}_n = \omega_n \mathbf{A}_n$. For orthonormal modes it then follows that $\langle \mathbf{A}_m | \hat{\omega} \mathbf{A}_n \rangle = \omega_n \delta_{n,m}$ so that the Hamiltonian is reduced to

$$H = \sum_n a_n^\dagger a_n \hbar \omega_n . \tag{3.26}$$

3.1.1 General construction of single-photon states

From the above it is clear that the annihilation operator for a specific mode function \mathbf{A}_n can be written as

$$\begin{aligned}
a_n &= \langle \mathbf{A}_n | \hat{\mathbf{A}} \rangle \\
&= \frac{i\varepsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot \dot{\hat{\mathbf{A}}} - \dot{\mathbf{A}}_n^* \cdot \hat{\mathbf{A}} \right) \\
&= \frac{i\varepsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot (-i\hat{\omega} \hat{\mathbf{A}}^{(+)} + i\hat{\omega} \hat{\mathbf{A}}^{(-)}) - i\hat{\omega} \mathbf{A}_n^* \cdot (\hat{\mathbf{A}}^{(+)} + \hat{\mathbf{A}}^{(-)}) \right) \\
&= \frac{i\varepsilon_0}{\hbar} \int d^3x \left(\mathbf{A}_n^* \cdot (-i\hat{\omega} \hat{\mathbf{A}}^{(+)} + i\hat{\omega} \hat{\mathbf{A}}^{(-)}) - i\mathbf{A}_n^* \cdot (\hat{\omega} \hat{\mathbf{A}}^{(+)} + \hat{\omega} \hat{\mathbf{A}}^{(-)}) \right) \\
&= \frac{2\varepsilon_0}{\hbar} \int d^3x \mathbf{A}_n^* \cdot \hat{\omega} \hat{\mathbf{A}}^{(+)} .
\end{aligned} \tag{3.27}$$

Now, $\hat{\mathbf{E}}^{(+)} = -\partial_t \hat{\mathbf{A}}^{(+)} = i\hat{\omega} \hat{\mathbf{A}}^{(+)}$, so that

$$a_n = \frac{2\epsilon_0}{i\hbar} \int d^3x \mathbf{A}_n^* \cdot \hat{\mathbf{E}}^{(+)} . \quad (3.28)$$

This means that by specifying the properly normalized field mode $\mathbf{A}_n(\mathbf{x}, t)$ we can construct the corresponding annihilation operator using the positive-frequency part of the electric field operator.

A single photon in this mode is described by the state

$$|\psi\rangle = a_n^\dagger |0\rangle . \quad (3.29)$$

If this photon interacts with an atom in the ground state $|g\rangle$, the latter will be excited and the photon will be absorbed at the position \mathbf{x}_0 of the atom. The corresponding interaction Hamiltonian can be written as $-\mathbf{d} \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{x}_0)|e\rangle\langle g| + \text{H.c.}$, so that the Rabi frequency of a single photon becomes

$$\begin{aligned} \hbar\Omega &= \langle 0| \otimes \langle e| \mathbf{d} \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{x}_0) |e\rangle \langle g| a_n^\dagger |0\rangle \otimes |g\rangle \\ &= \mathbf{d} \cdot \langle 0| \hat{\mathbf{E}}^{(+)}(\mathbf{x}_0) a_n^\dagger |0\rangle \\ &= \mathbf{d} \cdot [\hat{\mathbf{E}}^{(+)}(\mathbf{x}_0), a_n^\dagger] . \end{aligned} \quad (3.30)$$

The commutator can be evaluated by

$$\begin{aligned} [\hat{\mathbf{E}}_i^{(+)}(\mathbf{x}_0), a_n^\dagger] &= [\hat{\mathbf{E}}_i^{(+)}(\mathbf{x}_0), \frac{2\epsilon_0}{-i\hbar} \int d^3x \mathbf{A}_n(\mathbf{x}) \cdot \hat{\mathbf{E}}^{(-)}(\mathbf{x})] \\ &= \frac{2\epsilon_0}{-i\hbar} \int d^3x \mathbf{A}_{n,j}(\mathbf{x}) [\hat{\mathbf{E}}_i^{(+)}(\mathbf{x}_0), \hat{\mathbf{E}}_j^{(-)}(\mathbf{x})] \\ &\stackrel{(i)}{=} \frac{2\epsilon_0}{-i\hbar} \int d^3x \mathbf{A}_{n,j}(\mathbf{x}) \frac{\hbar\hat{\omega}}{2\epsilon_0} \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}_0) \\ &= i \int d^3x (\hat{\omega} \mathbf{A}_{n,j}(\mathbf{x})) \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}_0) \\ &\stackrel{(ii)}{=} i\hat{\omega} \mathbf{A}_{n,i}(\mathbf{x}_0) \\ &= -\partial_t \mathbf{A}_{n,i}(\mathbf{x}_0) \\ &= \mathbf{E}_{n,i}(\mathbf{x}_0) \end{aligned}$$

In step (i) we have used the fundamental commutation relation

$$[\hat{\mathbf{E}}_i^{(+)}(\mathbf{x}_0), \hat{\mathbf{E}}_j^{(-)}(\mathbf{x})] = \frac{\hbar\hat{\omega}}{2\epsilon_0} \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}_0) . \quad (3.31)$$

Step (ii) follows from the fact that the mode function \mathbf{A}_n is transverse. For any transverse vector field the transverse delta distribution acts like the ordinary delta distribution:

$$\int d^3x \mathbf{A}_{n,j}(\mathbf{x}) \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}_0) = \mathbf{A}_{n,i}(\mathbf{x}_0) . \quad (3.32)$$

We thus have found the result that the electric field of a single photon in mode \mathbf{A}_n is simply given by $\mathbf{E}_n = -\dot{\mathbf{A}}_n$. While this may appear self-evident it is an important check that the mode formalism is working correctly.

3.1.2 Gaussian modes in paraxial approximation

In this Section we will derive explicit expressions for normalized mode functions in the paraxial approximation that has been introduced in Sec. 1.8. However, since we are dealing with the frequency operator we will first rederive the paraxial approximation.

The paraxial approximation relies on the assumption that the field mode is almost monochromatic. In other words, its Fourier transform is narrowly peaked around a certain wavevector \mathbf{k}_L , which we will take to be $\mathbf{k}_L = k_L \mathbf{e}_z$. This is essentially the same statement as assuming that the field \mathbf{E}_n can be written as

$$\mathbf{E}_n = \mathcal{E}_n(\mathbf{x}, t) e^{ik_L z - i\omega_L t}, \quad \omega_L = ck_L, \quad (3.33)$$

where $\mathcal{E}_n(\mathbf{x}, t)$ is slowly varying both in space and in time so that $|\partial_z \mathcal{E}_n| \ll k_L |\mathcal{E}_n|$ and $|\partial_t \mathcal{E}_n| \ll \omega_L |\mathcal{E}_n|$. An immediate consequence of this is that

$$\begin{aligned} \nabla \cdot \mathbf{E}_n &= 0 \\ &= (\nabla \cdot \mathcal{E}_n + i\mathbf{k}_L \cdot \mathcal{E}_n) e^{ik_L z - i\omega_L t} \\ &\approx i\mathbf{k}_L \cdot \mathcal{E}_n e^{ik_L z - i\omega_L t}, \end{aligned} \quad (3.34)$$

so that we can write $\mathcal{E}_n(\mathbf{x}, t) \approx \epsilon \mathcal{E}_n(\mathbf{x}, t)$, where ϵ is a constant polarization vector orthogonal to \mathbf{k}_L .

Being a positive-frequency mode of the free electromagnetic field, \mathbf{E}_n fulfills the exact equation $i\partial_t \mathbf{E}_n = \hat{\omega} \mathbf{E}_n$. In Fourier space the right-hand side becomes ³

$$\begin{aligned} \hat{\omega} \mathbf{E}_n(\mathbf{x}) &= \hat{\omega} \int \frac{d^3 k}{(2\pi)^3} e^{i\mathbf{k} \cdot \mathbf{x}} \mathbf{E}_n(\mathbf{k}) \\ &= \int \frac{d^3 k}{(2\pi)^3} c|\mathbf{k}| e^{i\mathbf{k} \cdot \mathbf{x}} \mathbf{E}_n(\mathbf{k}), \end{aligned} \quad (3.35)$$

with

$$\begin{aligned} \mathbf{E}_n(\mathbf{k}) &= \int d^3 x' e^{-i\mathbf{k} \cdot \mathbf{x}'} \mathbf{E}_n(\mathbf{x}') \\ &= \int d^3 x' e^{-i\mathbf{k} \cdot \mathbf{x}'} \epsilon \mathcal{E}_n(\mathbf{x}') e^{i\mathbf{k}_L \cdot \mathbf{x}' - i\omega_L t} \\ &= \epsilon e^{-i\omega_L t} \int d^3 x' e^{-i\Delta \mathbf{k} \cdot \mathbf{x}'} \mathcal{E}_n(\mathbf{x}') \\ &= \epsilon e^{-i\omega_L t} \mathcal{E}_n(\Delta \mathbf{k}) \end{aligned} \quad (3.36)$$

³We suppress the time dependence to keep the notation simple.

with $\Delta \mathbf{k} \equiv \mathbf{k} - \mathbf{k}_L$. Since by assumption $\mathbf{E}_n(\mathbf{k})$ is narrowly peaked around \mathbf{k}_L we can expand the frequency that appears in Eq. (3.35) as

$$\begin{aligned} c|\mathbf{k}| &= c\sqrt{\mathbf{k}^2} \\ &= c\sqrt{(\mathbf{k}_L + \Delta \mathbf{k})^2} \\ &= c\sqrt{k_L^2 + 2k_L\Delta k_z + \Delta k_z^2 + \Delta \mathbf{k}_x^2 + \Delta \mathbf{k}_y^2}. \end{aligned} \quad (3.37)$$

Since $|\Delta \mathbf{k}| \ll k_L$ we can expand the square root in lowest order in the components of $\Delta \mathbf{k}$ to obtain

$$c|\mathbf{k}| \approx ck_L + c\Delta k_z + \frac{c}{2k_L}(\Delta \mathbf{k}_x^2 + \Delta \mathbf{k}_y^2). \quad (3.38)$$

Thus,

$$\begin{aligned} \hat{\omega} \mathbf{E}_n(\mathbf{x}) &= \epsilon e^{-i\omega_L t} \int \frac{d^3 k}{(2\pi)^3} c|\mathbf{k}| e^{i\mathbf{k} \cdot \mathbf{x}} \mathcal{E}_n(\Delta \mathbf{k}) \\ &= \epsilon e^{ik_L z - i\omega_L t} \int \frac{d^3 \Delta \mathbf{k}}{(2\pi)^3} c|\mathbf{k}_L + \Delta \mathbf{k}| e^{i\Delta \mathbf{k} \cdot \mathbf{x}} \mathcal{E}_n(\Delta \mathbf{k}) \\ &\approx \epsilon e^{ik_L z - i\omega_L t} \int \frac{d^3 \Delta \mathbf{k}}{(2\pi)^3} \left(ck_L + c\Delta k_z + \frac{c}{2k_L} (\Delta \mathbf{k}_x^2 + \Delta \mathbf{k}_y^2) \right) e^{i\Delta \mathbf{k} \cdot \mathbf{x}} \mathcal{E}_n(\Delta \mathbf{k}) \\ &\approx \epsilon e^{ik_L z - i\omega_L t} \left(ck_L - ic\partial_z - \frac{c}{2k_L} (\partial_x^2 + \partial_y^2) \right) \mathcal{E}_n(\mathbf{x}) \end{aligned} \quad (3.39)$$

This corresponds to the paraxial approximation. The corresponding paraxial wave equation follows by inserting this in the dynamic equation,

$$\begin{aligned} (i\partial_t - \hat{\omega}) \mathbf{E}_n &= \epsilon e^{ik_L z - i\omega_L t} (\omega_L \mathcal{E}_n + i\partial_t \mathcal{E}_n) - \hat{\omega} \mathbf{E}_n \\ &\approx \epsilon e^{ik_L z - i\omega_L t} \left(\omega_L + i\partial_t - ck_L + ic\partial_z + \frac{c}{2k_L} (\partial_x^2 + \partial_y^2) \right) \mathcal{E}_n \end{aligned} \quad (3.40)$$

so that

$$\boxed{(i\partial_t - \hat{\omega}) \epsilon e^{ik_L z - i\omega_L t} \mathcal{E}_n \approx \epsilon e^{ik_L z - i\omega_L t} \left(i(\partial_t + c\partial_z) + \frac{c}{2k_L} (\partial_x^2 + \partial_y^2) \right) \mathcal{E}_n}. \quad (3.41)$$

In Sec. 1.8 we have studied monochromatic solutions to these equations which provide an excellent description of a laser beam. However, monochromatic fields are not normalizable so that they are not useful to describe the state of a single photon. To get normalizable states we need to admit pulses. To find such states

we can exploit the close similarity of the paraxial wave equation and the free Schrödinger equation. It is then easy to see that a Gaussian mode is given by

$$\mathcal{E} = \frac{E_{\max} w_0^2}{\eta(t)} e^{-\frac{x^2+y^2}{2\eta(t)}} e^{-\frac{(-c(t-t_0)+z-z_0)^2}{2c^2\tau^2}} , \quad \eta(t) \equiv w_0^2 + i \frac{c(t-t_0)}{k_L} . \quad (3.42)$$

In this expression τ denotes the pulse duration which corresponds to the time in which the intensity $I = 2\varepsilon_0 c |\mathcal{E}|^2$ (or rather the exponential factor that appears in I) drops to $1/e$ of its maximum value. Likewise, w_0 denotes the transverse $1/e$ width of I for $t = t_0$ and $z = z_0$. E_{\max} is the maximum electric field in this mode and can be calculated by normalizing the mode,

$$\begin{aligned} \langle \mathbf{A}_n | \mathbf{A}_n \rangle &= \frac{2\varepsilon_0}{\hbar} \int d^3x \mathbf{E}_n^* \cdot \hat{\omega}^{-1} \mathbf{E}_n \\ &= \frac{2\varepsilon_0}{\hbar\omega_L} \int d^3x |\mathbf{E}_n|^2 \\ &= \frac{2\varepsilon_0}{\hbar\omega_L} \int d^3x |\mathcal{E}_n|^2 \\ &= \frac{2\varepsilon_0 \tau E_{\max}^2 \pi^{3/2} w_0^2}{\hbar k_L} , \end{aligned} \quad (3.43)$$

so that

$$E_{\max} = \sqrt{\frac{\hbar k_L}{2\varepsilon_0 \tau \pi^{3/2} w_0^2}} = \sqrt{\frac{\hbar\omega_L}{V \varepsilon_0 \pi^{1/2}}} \quad (3.44)$$

where $V = c 2\tau \pi w_0^2$ is the volume of the mode in vacuum. This is remarkably similar to the normalization factor of plane-wave modes if the electromagnetic field is quantized in a box.

3.2 Coherent States

Coherent states are probably the most important states in quantum optics and represent a quantum mechanical description of classical fields.

Consider a single mode a , $[a, a^\dagger] = 1$ of the electromagnetic field. There are several definitions of coherent states $|\alpha\rangle$:

- Eigenstate of a : $a |\alpha\rangle = \alpha |\alpha\rangle$, $\alpha \in \mathbb{C}$
- state of minimal uncertainty (exercises)
- Displacement of the vacuum

What is $a|\alpha\rangle$? Expand $|\alpha\rangle$ in number states:

$$|\alpha\rangle = \sum_{n=0}^{\infty} \alpha_n |n\rangle = \sum_{n=0}^{\infty} \frac{\alpha_n}{\sqrt{n!}} (a^\dagger)^n |0\rangle$$

$$\Rightarrow a|\alpha\rangle = \sum_{n=0}^{\infty} \alpha_n a |n\rangle = \sum_{n=1}^{\infty} \alpha_n \sqrt{n} |n-1\rangle$$

With $n' = n - 1$ one finds

$$a|\alpha\rangle = \sum_{n'=0}^{\infty} \alpha_{n'+1} \sqrt{n'+1} |n'\rangle \stackrel{!}{=} \alpha|\alpha\rangle = \alpha \sum_{n'=0}^{\infty} \alpha_{n'} |n'\rangle$$

It follows that $\alpha_{n+1} = \alpha \frac{\alpha_n}{\sqrt{n+1}}$, whereby α_0 has to be fixed by the normalization condition. From

$$\begin{aligned} \alpha_1 &= \alpha \alpha_0 \\ \alpha_2 &= \frac{\alpha^2}{\sqrt{2!}} \alpha_0 \\ \alpha_3 &= \frac{\alpha^3}{\sqrt{3!}} \alpha_0 \text{ etc.} \end{aligned}$$

one can deduce

$$\alpha_n = \frac{\alpha^n}{\sqrt{n!}} \alpha_0 \quad (3.45)$$

This results in

$$|\alpha\rangle = \alpha_0 \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle = \alpha_0 \sum_{n=0}^{\infty} \frac{\alpha^n}{n!} (a^\dagger)^n |0\rangle = \alpha_0 \exp(\alpha a^\dagger) |0\rangle$$

Using $\langle n|m\rangle = \delta_{nm}$ and Eq. (3.45) one is led to

$$\langle \alpha|\alpha\rangle = 1 = |\alpha_0|^2 \sum_{n=0}^{\infty} \frac{(\alpha^*)^n}{\sqrt{n!}} \langle n| \sum_{m=0}^{\infty} \frac{\alpha^m}{\sqrt{m!}} |m\rangle = |\alpha_0|^2 \sum_{n=0}^{\infty} \frac{|\alpha|^{2n}}{n!} = |\alpha_0|^2 \exp |\alpha|^2 \stackrel{!}{=} 1$$

This leads to

$$\alpha_0 = \exp\left(-\frac{|\alpha|^2}{2}\right) \quad (3.46)$$

We thus can represent the coherent state $|\alpha\rangle$ as

$$\boxed{|\alpha\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right) \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right) \exp(\alpha a^\dagger) |0\rangle} \quad (3.47)$$

$|\alpha\rangle$ can also be constructed using the *displacement operator*

$$\boxed{D(\alpha) = \exp(\alpha a^\dagger - \alpha^* a)} . \quad (3.48)$$

To do so we can use the Baker-Campbell-Hausdorff equation:

$$e^{(A+B)} = e^A e^B e^{-\frac{1}{2}[A,B]} e^{-\frac{1}{6}[A,[A,B]]} \dots \quad (3.49)$$

Inserting $A = \alpha a^\dagger$ and $B = -\alpha^* a$ we find

$$\begin{aligned} -\frac{1}{2}[A, B] &= -\frac{1}{2}(-|\alpha|^2)[a^\dagger, a] = -\frac{|\alpha|^2}{2} \in \mathbb{R} \\ \Rightarrow [A, [A, B]] &= 0 \end{aligned}$$

Together with Eq. (3.49) the then can be brought into the form

$$\boxed{D(\alpha) = e^{\alpha a^\dagger} e^{-\alpha^* a} e^{-\frac{|\alpha|^2}{2}}} \quad (3.50)$$

This allows us to prove the following properties of the displacement operator:

- $D(\alpha)$ is unitary:

$$D^\dagger(\alpha) = e^{\alpha^* a - \alpha a^\dagger} = e^{-(\alpha a^\dagger - \alpha^* a)} = D^{-1}(\alpha) = D(-\alpha)$$

- We have the relation

$$|\alpha\rangle = D(\alpha) |0\rangle \quad (3.51)$$

It becomes clear from Eq. (3.51) that the vacuum can be considered as the special coherent state $\alpha = 0$.

It was shown in the excercises that ⁴

$$E \sim a + a^\dagger \quad (3.52)$$

$$B \sim i(a^\dagger - a) \quad (3.53)$$

Thus,

$$\bar{E} = \langle \alpha | E | \alpha \rangle = \langle \alpha | a + a^\dagger | \alpha \rangle = \alpha + \alpha^* = 2\text{Re } \alpha \quad (3.54)$$

$$\bar{B} = \langle \alpha | B | \alpha \rangle = \langle \alpha | i(a^\dagger - a) | \alpha \rangle = i(\alpha^* - \alpha) = 2\text{Im } \alpha \quad (3.55)$$

⁴Strictly speaking this holds only for a certain choice of polarization vector and phase. For a single mode we have $\mathbf{A} \propto a \exp(ikx)\boldsymbol{\epsilon} + a^\dagger \exp(-ikx)\boldsymbol{\epsilon}^*$ and therefore $\mathbf{E} = -\dot{\mathbf{A}} \propto i\omega a \exp(ikx)\boldsymbol{\epsilon} + (-i\omega)a^\dagger \exp(-ikx)\boldsymbol{\epsilon}^*$ as well as $\mathbf{B} = \text{curl } \mathbf{A} \propto ia \exp(ikx)\mathbf{k} \times \boldsymbol{\epsilon} + (-i)a^\dagger \exp(-ikx)\mathbf{k} \times \boldsymbol{\epsilon}^*$. Choosing, for instance, circularly polarized light ($\boldsymbol{\epsilon} \propto \mathbf{e}_x + i\mathbf{e}_y$) and $x = 0$ one finds that the y components of the fields fullfill the above relations.

$$\begin{aligned}
(\Delta E)^2 &= \langle E^2 \rangle - \bar{E}^2 \\
&= \langle \alpha | a^2 + aa^\dagger + a^\dagger a + (a^\dagger)^2 | \alpha \rangle - (\alpha - \alpha^*)^2 \\
&= \alpha^2 + (\alpha^*)^2 + \alpha^* \alpha + \langle \alpha | aa^\dagger | \alpha \rangle - (\alpha - \alpha^*)^2 \\
&= \alpha^2 + (\alpha^*)^2 + \alpha^* \alpha + \langle \alpha | a^\dagger a + [a, a^\dagger] | \alpha \rangle - (\alpha - \alpha^*)^2 \\
&= \alpha^2 + (\alpha^*)^2 + \alpha^* \alpha + \langle \alpha | a^\dagger a + 1 | \alpha \rangle - (\alpha - \alpha^*)^2 \\
&= \alpha^2 + (\alpha^*)^2 + 2\alpha\alpha^* + 1 - \alpha^2 - (\alpha^*)^2 - 2\alpha\alpha^* \\
&= 1
\end{aligned} \tag{3.56}$$

$\Rightarrow \Delta E$ is independent from α . Analogous calculation for ΔB :

$$(\Delta B)^2 = \dots = 1 \tag{3.57}$$

$$\Rightarrow \Delta E \Delta B = 1 \tag{3.58}$$

The general form of Heisenberg's uncertainty relation for two (arbitrary) hermitean operators is given by $\Delta E \Delta B \geq |\langle [E, B] \rangle|/2$. From this we can see that coherent states have minimum uncertainty.

\Rightarrow One can characterize coherent states by a complex amplitude $\alpha \in \mathbb{C}$ and by an uncertainty 1. This translates into the following graphical picture

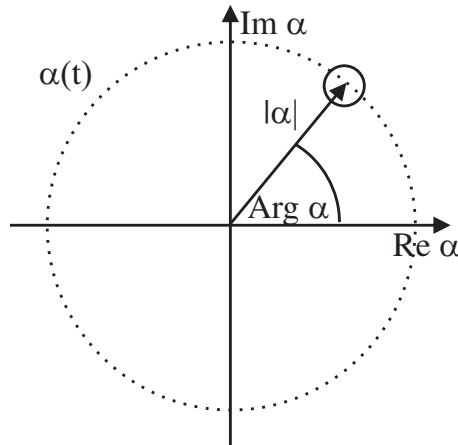


Figure 3.1: Visualization of a coherent state by a complex amplitude with uncertainty 1 (small circle). The dashed circle describes the (free) time evolution of the amplitude.

Free time evolution of coherent states:

$$|\alpha(t)\rangle = U(t) |\alpha(0)\rangle \propto U e^{\alpha a^\dagger} |0\rangle = U e^{\alpha a^\dagger} U^\dagger U |0\rangle$$

$$U|0\rangle = |0\rangle \Rightarrow |\alpha(t)\rangle = Ue^{\alpha a^\dagger}U^\dagger|0\rangle$$

$$\begin{aligned} Ue^A U^\dagger &= U(1 + A + \frac{A^2}{2} + \dots)U^\dagger \\ &= UU^\dagger + UAU^\dagger + \frac{1}{2}UA(U^\dagger U)AU^\dagger + \dots \\ &= 1 + UAU^\dagger + \frac{1}{2}(UAU^\dagger)^2 + \dots \\ &= \exp(UAU^\dagger) \end{aligned} \quad (3.59)$$

$$\Rightarrow Ue^{\alpha a^\dagger}U^\dagger = e^{\alpha Ua^\dagger U^\dagger} = \exp(\alpha e^{-i\omega t}a^\dagger) \quad (3.60)$$

$$\Rightarrow |\alpha(t)\rangle = |\alpha e^{-i\omega t}\rangle \quad (3.61)$$

Hence, coherent states remain coherent states and their amplitude moves on a circle around the origin of the complex plane.

- Coherent states are *not orthogonal* to each other:

$$\langle\alpha|\beta\rangle = e^{\frac{-|\alpha|^2}{2}} e^{\frac{-|\beta|^2}{2}} e^{\alpha^*\beta} \quad (3.62)$$

The proof can be done using the expansion in number states or the displacement operator and Baker-Campbell-Hausdorff equation (Eq. (3.49) on page 53).

- Coherent states are *overcomplete*:

$$\int d^2\alpha |\alpha\rangle \langle\alpha| = \pi \mathbf{1} \quad (3.63)$$

Proof: Expansion in number states and integration over the phase angle of α .

3.3 Classical and Quantum Mechanical Interference

The difference between classical and quantum mechanical interference is at first sight rather subtle, but can be well understood using coherent states. Set $|\psi\rangle = |\alpha\rangle = D(\alpha)|0\rangle$; $D^\dagger(\alpha) = D^{-1}(\alpha)$.

$D(\alpha)$ can be used to switch to a unitarily equivalent „picture“ in which the state $|\tilde{\psi}\rangle = D^{-1}(\alpha)|\psi\rangle$ coincides with the vacuum. The operators then transform as

$$\hat{\tilde{O}} = D^\dagger(\alpha)\hat{O}D(\alpha) \quad (3.64)$$

$$(\Longleftrightarrow \langle \tilde{\psi} | \hat{O} | \tilde{\psi} \rangle = \langle \psi | \hat{O} | \psi \rangle)$$

$$\begin{aligned} \tilde{\mathbf{E}}(x) &= D^\dagger(\alpha) \mathbf{E}(x) D(\alpha) \\ &= \sum_{a_{k\sigma} \neq a} (a_{k\sigma} \dots e^{i\mathbf{k}\mathbf{x}} + \text{H.c.}) + i\sqrt{\frac{\hbar\omega_a}{2(2\pi)^3\epsilon_0}} \epsilon_a e^{i\mathbf{k}\mathbf{x}} D^\dagger(\alpha) a D(\alpha) + \text{H.c.} \\ &= \int dk \sum_{\sigma} a_{k\sigma} \dots + \text{H.c.} + i\sqrt{\dots} \epsilon_a e^{i\mathbf{k}\mathbf{x}} D^\dagger a D + \text{H.c.} \end{aligned}$$

To calculate $D^\dagger(\alpha) a D(\alpha)$ we use the theorem

$$\boxed{e^A B e^{-A} = \sum_{n=0}^{\infty} \frac{1}{n!} k_n} \quad k_0 = B; \quad k_{n+1} = [A, k_n] \quad (3.65)$$

$$\begin{aligned} \implies A &= \alpha^* a - \alpha a^\dagger; B = a \\ \implies k_0 &= a; k_1 = [\alpha^* a - \alpha a^\dagger, a] = \alpha \\ \implies k_2 &= k_3 = \dots = 0 \\ \implies \boxed{D^\dagger(\alpha) a D(\alpha) = a + \alpha} \end{aligned}$$

$$\begin{aligned} \tilde{\mathbf{E}}(x) &= \mathbf{E}_{\text{All other modes}} + (i\sqrt{\dots} \epsilon_a e^{i\mathbf{k}\mathbf{x}} a - i\sqrt{\dots} \epsilon_a^* e^{-i\mathbf{k}\mathbf{x}} a^\dagger) \\ &\quad + (i\alpha\sqrt{\dots} \epsilon_a e^{i\mathbf{k}\mathbf{x}} - i\sqrt{\dots} \epsilon_a^* \alpha^* e^{-i\mathbf{k}\mathbf{x}}) \\ \implies \boxed{\tilde{\mathbf{E}}(x) = \mathbf{E}(x) + \mathbf{E}_a^{\text{classical}}(x)} \end{aligned} \quad (3.66)$$

The state $|\alpha\rangle$ in absence of external classical fields is thus unitarily equivalent to the vacuum in the presence of the field $\mathbf{E}_a^{\text{class.}}(x)$ (and $\mathbf{B}_a^{\text{class.}}(x)$).

We can exploit this equivalence to understand the difference between the interference of photons and classical Maxwell fields:

- Classical interference:

$$\mathbf{E} = \mathbf{E}_1 + \mathbf{E}_2, \quad \text{e.g. } \mathbf{E}_i \propto e^{i\mathbf{k}_i \mathbf{x}}$$

$$\begin{aligned} I \propto \mathbf{E} \mathbf{E}^* &= |\mathbf{E}_1|^2 + |\mathbf{E}_2|^2 + 2\text{Re}(\mathbf{E}_1 \mathbf{E}_2^*) \\ &= 2\text{Re}(\mathbf{E}_1 \mathbf{E}_2^*) \propto \cos(\mathbf{x}(\mathbf{k}_1 - \mathbf{k}_2)) \end{aligned}$$

- Quantum description of classical interference:

$$\begin{aligned}
 |\psi\rangle &= |\alpha_1, \alpha_2\rangle \\
 &= D_1(\alpha_1)D_2(\alpha_2)|0\rangle \\
 D_1(\alpha_1) &= e^{\alpha_1 a_1^\dagger - \alpha_1^* a_1} \\
 D_2(\alpha_2) &\quad \text{analogously}
 \end{aligned}$$

The product of operators $D_1 D_2$ creates the sum of classical fields,

$$D_2^\dagger D_1^\dagger \hat{\mathbf{E}} D_1 D_2 = D_2^\dagger (\hat{\mathbf{E}} + \mathbf{E}_1) D_2 = \hat{\mathbf{E}} + \mathbf{E}_1 + \mathbf{E}_2$$

Transformation using $D_2^\dagger D_1^\dagger$ from $|\psi\rangle$ to the vacuum:

$$\begin{aligned}
 I \propto \langle \psi | \mathbf{E}^{(-)} \mathbf{E}^{(+)} | \psi \rangle &= \langle 0 | (\hat{\mathbf{E}}^{(-)} + \mathbf{E}_1^* + \mathbf{E}_2^*) (\hat{\mathbf{E}}^{(+)} + \mathbf{E}_1 + \mathbf{E}_2) | 0 \rangle \\
 &= |\mathbf{E}_1 + \mathbf{E}_2|^2 \\
 &\iff \text{classical interference fringes}
 \end{aligned}$$

In quantum mechanics one can also consider superpositions of coherent states. This does not describe classical fields anymore:

$$|\psi\rangle = |\alpha_1\rangle + |\alpha_2\rangle = (D_1(\alpha_1) + D_2(\alpha_2))|0\rangle$$

Interference fringes for this state:

$$\begin{aligned}
 I &\propto \langle \psi | \hat{\mathbf{E}}^{(-)} \hat{\mathbf{E}}^{(+)} | \psi \rangle \\
 &= \langle 0 | (D_1^\dagger + D_2^\dagger) \hat{\mathbf{E}}^{(-)} \hat{\mathbf{E}}^{(+)} (D_1 + D_2) | 0 \rangle \\
 &= \langle 0 | (D_1^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_1 + D_1^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_2 + D_2^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_1 + D_2^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_2) | 0 \rangle \\
 &= |\mathbf{E}_1|^2 + |\mathbf{E}_2|^2 + \langle 0 | (D_1^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_2 + D_2^\dagger \mathbf{E}^{(-)} \mathbf{E}^{(+)} D_1) | 0 \rangle \\
 &= |\mathbf{E}_1|^2 + |\mathbf{E}_2|^2 + \langle \alpha_1 | \mathbf{E}^{(-)} \mathbf{E}^{(+)} | \alpha_2 \rangle + \langle \alpha_2 | \mathbf{E}^{(-)} \mathbf{E}^{(+)} | \alpha_1 \rangle
 \end{aligned}$$

$$\begin{aligned}
 \implies I &\propto |\mathbf{E}_1|^2 + |\mathbf{E}_2|^2 + E_1^* E_2 \langle \alpha_1 | \alpha_2 \rangle + E_2^* E_1 \langle \alpha_2 | \alpha_1 \rangle \\
 \langle \alpha_1 | \alpha_2 \rangle &= e^{-\frac{|\alpha_1|^2}{2}} e^{-\frac{|\alpha_2|^2}{2}} e^{\alpha_1^* \alpha_2} \\
 \implies |\langle \alpha_1 | \alpha_2 \rangle|^2 &= \dots = e^{-|\alpha_1 - \alpha_2|^2}
 \end{aligned}$$

\implies For $|\alpha_1 - \alpha_2| \gg 1$ quantum interference is strongly suppressed. For a classical field with a large number of photons $|\alpha|^2 \gg 1$ it is therefore unimportant. The state $|\alpha_1\rangle + |\alpha_2\rangle$ is also called Schrödinger cat state since it corresponds to a superposition of macroscopic states (for $|\alpha_i|^2 \gg 1$).

3.4 Linear optical elements

The mode expansion given above clearly indicates that a complete quantum theory of light must contain all possible modes of the electromagnetic field. However, in certain situations that are quite common in quantum optics experiments it is possible to ignore all but a few or even a single mode. The reason behind this is that the free wave equation is linear so that superpositions of mode functions again correspond to a mode function. Hence, if we do not observe the shape of the mode function but only explore the quantum statistical properties of light (e.g., by counting the clicks of a detector) we may assume that the photons always occupy one specific mode whose annihilation operator a corresponds to a single harmonic oscillator.

There are of course a number of conditions when this approximation can be applied. Obviously the mode functions of all photons involved must almost perfectly overlap. We therefore need a photon source that always generates photons in nearly the same mode. The Hong-Ou-Mandel dip (see below) demonstrates what can happen if this condition is not fulfilled.

Furthermore, the single mode approximation can break down if light interacts with a medium in such a way that photons are distorted in a different way. This is in particular the case in nonlinear media where the presence of other photons distorts the mode of a photon. In that case the assumption that all photons occupy a given mode clearly breaks down. It may even happen that the notion of a single-photon mode becomes meaningless because genuine two-photon states (e.g., more complicated entangled states) are needed for a proper description.

Despite these restrictions the single- (or few-)mode approximation is extremely useful if only linear optical elements are used to manipulate photons. Ignoring the dynamics of the mode then allows to describe the quantum aspects of photons with a very simple harmonic oscillator model that is tremendously helpful to sharpen the intuition about quantum mechanics.

In this section we will discuss a simple description of lossless linear optical elements in the context of the few-mode approximation. Linear optical elements include phase shifters, polarizing and non-polarizing beam splitters, and polarization rotators (e.g., quarter-wave plates).

3.4.1 The basic principles of lossless linear optical elements

Lossless passive linear optical elements are characterized by three features:

- Absence of losses: this is an idealization that works quite well for linear optical elements. It states that no photons are lost when light passes through

the element. In quantum mechanics this translates into the unitarity of the evolution.

- **Passivity:** there is no source of energy in these elements that could change the energy of the light field. In other words, the energy of the incoming light beam is conserved. Furthermore, the usual case in quantum optics is that the frequency of the light is not changed either. energy conservation then implies that the total number of photons in passive linear optical elements is conserved.
- **Linearity:** there are no nonlinear effects associated with an optical element. This means the evolution of the quantum state does not depend on the photon number or the intensity of the light beam.

Taken together these conditions imply that lossless passive linear optical elements may be described by a unitary transformation U that conserves photon number and energy. One can phenomenologically construct U by making a suitable ansatz for an effective Hamiltonian inside the medium and calculate the corresponding time evolution. In the following we will do this for a number of common elements.

3.4.2 Phase shifter

A phase shifter can simply be a piece of dielectric material like glass that can change the phase of a light beam relative to the phase it acquires during free evolution. In practise, however, things are more complicated ⁵: one often wants to change the phase continuously which cannot be accomplished easily with a piece of glass. Instead, a mirror is fixed on a piezo crystal and put somewhere into the path of the light beam. Applying a voltage to the crystal then allows to change the position of the mirror by very small amounts. The resulting change in the optical path's length then generates a phase shift.

For simplicity, and to motivate the theoretical description for the phase shifter given below, we stick to a model in which the phase shift is generated by a dielectric medium that slightly shifts the frequency of light. The dynamical phase factor for light propagating for a time t is given by $\exp(-i\omega t)$ where t is the duration the light is inside the phase plate ⁶. If the frequency of light inside the phase plate is slightly different, say ω' , then the effect of the medium would be to generate

⁵I thank Alex Lvovsky for pointing this out to me.

⁶Actually the frequency remains constant but the speed of light is changed so that the time t needed to propagate through the dielectric, but for the sake of developing a unified phenomenological picture of linear optical elements we deal with the medium as if it was changing the frequency

a relative phase shift of $\exp(-i\Delta\omega t)$ with $\Delta\omega = \omega' - \omega$. To construct the unitary transformation U_{PS} corresponding to a phase shifter we therefore assume an effective interaction Hamiltonian of the form

$$H_{\text{PS}} = \hbar\Delta\omega a^\dagger a . \quad (3.67)$$

The unitary transformation $U_{\text{PS}} = \exp(-itH_{\text{PS}}/\hbar)$ then becomes

$$U_{\text{PS}} = \exp(i\phi a^\dagger a) \quad (3.68)$$

with $\phi = -\Delta\omega t$.

Let us explore the effect of this operator on a general quantum state $|\psi\rangle = \sum_{n=0}^{\infty} \psi_n |n\rangle$ of the single-mode harmonic oscillator. Because $|n\rangle = (a^\dagger)^n |0\rangle / \sqrt{n!}$ we can express this state also as some function $f(a^\dagger) = \sum f_n (a^\dagger)^n$ applied to the vacuum state. Therefore

$$\begin{aligned} U_{\text{PS}}|\psi\rangle &= U_{\text{PS}} \sum_{n=0}^{\infty} f_n (a^\dagger)^n |0\rangle \\ &= \sum_{n=0}^{\infty} f_n (U_{\text{PS}} a^\dagger U_{\text{PS}}^\dagger)^n U_{\text{PS}} |0\rangle . \end{aligned} \quad (3.69)$$

Now $U_{\text{PS}}|0\rangle = |0\rangle$, the problem of finding the effect of U_{PS} on a general state is therefore reduced to finding its effect on a^\dagger . This is given by the

Phase shifter rule:

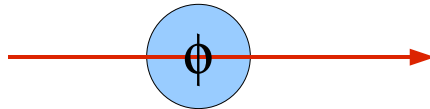
$$U_{\text{PS}} a U_{\text{PS}}^\dagger = e^{-i\phi} a \quad (3.70)$$

$$U_{\text{PS}} a^\dagger U_{\text{PS}}^\dagger = e^{i\phi} a^\dagger \quad (3.71)$$

Proof: for any two operators A, B one has

$$e^A B e^{-A} = \sum_{l=0}^{\infty} \frac{K_l}{l!} , \text{ with } K_0 = B , K_{l+1} = [A, K_l] \quad (3.72)$$

(this statement can be proven by calculating the Taylor expansion of $e^{xA} B e^{-xA}$ around $x = 0$). In our case we have $B = a$ and $A = i\phi a^\dagger a$, so $K_0 = a$ and $K_1 = i\phi [a^\dagger a, a] = -i\phi a$. Continuing this leads to $K_l = (-i\phi)^l a$ from which the first rule easily follows. The rule for a^\dagger is simply the adjoint of the rule for a .



Graphical representation of the phase shifter operation U_{PS}

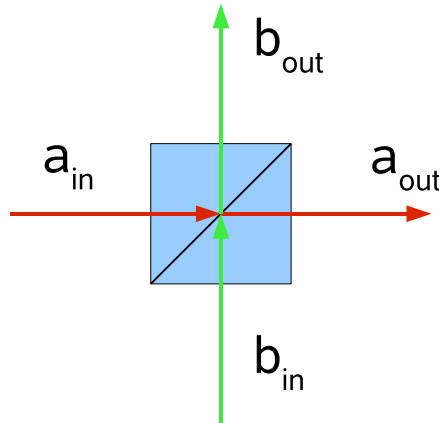
We have to check if this definition of the action of a phase shifter is in agreement with the observations for a classical beam of light. This means that, if the light is in a coherent state $|\alpha\rangle$, the mean (complex) amplitude of the electric field should exhibit a phase shift of ϕ . This is shown as follows,

$$\begin{aligned}
 U_{\text{PS}}|\alpha\rangle &= U_{\text{PS}} \exp(\alpha a^\dagger - \alpha^* a)|0\rangle \\
 &= U_{\text{PS}} \exp(\alpha a^\dagger - \alpha^* a) U_{\text{PS}}^\dagger U_{\text{PS}}|0\rangle \\
 &= \exp(U_{\text{PS}}(\alpha a^\dagger - \alpha^* a)U_{\text{PS}}^\dagger) U_{\text{PS}}|0\rangle \\
 &= \exp(\alpha e^{i\phi} a^\dagger - \alpha^* a e^{-i\phi})|0\rangle
 \end{aligned} \tag{3.73}$$

Therefore the complex amplitude α of the coherent state is changed to $\alpha e^{i\phi}$ as expected. We remark that the step in which U_{PS} is moved into the argument of the exponential can easily be proven when one writes the exponential as a Taylor series.

3.4.3 Beam splitter

A beam splitter is a device that splits a classical light beam into two beams with a certain ratio of intensities. In quantum optics the analogous action would be that an incoming photon in mode a_{in} should be prepared in a coherent superposition of the two outgoing modes a_{out} and b_{out} . Because a beam splitter also does not change the total number of photons, unitarity dictates that we have to include a second input channel b_{in} to describe a beam splitter (see figure). The reason is that otherwise a single state $|1_{a_{\text{in}}}\rangle$ would be mapped to some superposition of two output states, which cannot be accomplished with a unitary transformation if different superpositions are possible.



Graphical representation of the beam splitter operation U_{BS}

There are two different conventions in the literature with respect to denoting operators for input and output channels. The notation used in the previous paragraph assigns different names to input and output channels. However, one can also avoid introducing a name for the output operators and express everything in terms of the input operators. This is the convention we will be using here.

To construct the unitary operator U_{BS} we devise a Hamiltonian that describes the transfer from a photon in mode a to mode b and vice versa. A phenomenological ansatz that preserves the total number of photons is given by

$$H_{\text{BS}} = \zeta b^\dagger a + \zeta^* a^\dagger b \quad (3.74)$$

Similarly as for the phase shifter we find the corresponding unitary operator to be

$$U_{\text{BS}} = \exp(\theta b^\dagger a - \theta^* a^\dagger b) . \quad (3.75)$$

In complete analogy to the phase shifter we can again infer that $U_{\text{BS}}|0\rangle = |0\rangle$ and that the transformation of a general state can be deduced from the transformation law of the creation operator. We use again Eq. (3.72) to find this.

$$U_{\text{BS}} a U_{\text{BS}}^\dagger = e^{\theta b^\dagger a - \theta^* a^\dagger b} a e^{-\theta b^\dagger a + \theta^* a^\dagger b} \quad (3.76)$$

We then have $K_0 = a$, $K_1 = \theta^* b$, and $K_2 = -|\theta|^2 a = -|\theta|^2 K_0$. It is then easy to see that $K_{2l} = (-|\theta|^2)^l a$ and $K_{2l+1} = (-|\theta|^2)^l \theta^* b$. If we sum the terms for even and odd l separately we can see that they correspond to the Taylor expansion of the cosine and sine function, respectively:⁷

$$\begin{aligned} U_{\text{BS}} a U_{\text{BS}}^\dagger &= \sum_{l=0}^{\infty} \frac{K_l}{l!} \\ &= \sum_{n=0}^{\infty} \frac{K_{2n}}{(2n)!} + \sum_{n=0}^{\infty} \frac{K_{2n+1}}{(2n+1)!} \\ &= \sum_{n=0}^{\infty} \frac{(-1)^n |\theta|^{2n}}{(2n)!} a + \sum_{n=0}^{\infty} \frac{(-1)^n |\theta|^{2n} \theta^*}{(2n+1)!} b \\ &= a \cos |\theta| + b \frac{\theta^*}{|\theta|} \sin |\theta| \end{aligned} \quad (3.77)$$

For all operators involved we find the following

⁷If polarization is important these rules should be applied for each polarization direction, see the remarks given for the polarizing beam splitter below.

Beam splitter rules:

$$U_{\text{BS}} a U_{\text{BS}}^\dagger = a \cos |\theta| + b \frac{\theta^*}{|\theta|} \sin |\theta| \quad (3.78)$$

$$U_{\text{BS}} b U_{\text{BS}}^\dagger = b \cos |\theta| - a \frac{\theta}{|\theta|} \sin |\theta| \quad (3.79)$$

$$U_{\text{BS}} a^\dagger U_{\text{BS}}^\dagger = a^\dagger \cos |\theta| + b^\dagger \frac{\theta}{|\theta|} \sin |\theta| \quad (3.80)$$

$$U_{\text{BS}} b^\dagger U_{\text{BS}}^\dagger = b^\dagger \cos |\theta| - a^\dagger \frac{\theta^*}{|\theta|} \sin |\theta| \quad (3.81)$$

These equations describe a general lossless beam splitter. If the input-output notation is used one simply identifies $a_{\text{out}} = U_{\text{BS}} a U_{\text{BS}}^\dagger$ and so on. If we apply these rules to a superposition of two classical light fields,

$$|\psi_{\text{in}}\rangle = D_a(\alpha) D_b(\beta) |0\rangle = \exp(\alpha a^\dagger - \alpha^* a) \exp(\beta b^\dagger - \beta^* b) |0\rangle \quad (3.82)$$

we find, in the same way as for the phase shifter, that the classical complex amplitudes transform as

$$\alpha_{\text{out}} = \alpha \cos |\theta| - \beta \frac{\theta^*}{|\theta|} \sin |\theta| \quad (3.83)$$

$$\beta_{\text{out}} = \beta \cos |\theta| + \alpha \frac{\theta}{|\theta|} \sin |\theta| \quad (3.84)$$

which exactly corresponds to the classical behaviour we would expect.

The modulus and phase of θ determine the reflection and transmission coefficients of the beam splitter and the phase shift between a reflected and transmitted wave, respectively. Clearly the transmission coefficient for the electric field amplitude is given by $T = \cos |\theta|$ and the reflection coefficient by $R = \sin |\theta|$. The phase shift is not treated consistently in the literature because in a real beam splitter the relative phase between a reflected and a transmitted wave is random (e.g., making the propagation length of these two beams inside the beam splitter equal to within one wavelength would be extremely costly). One therefore can choose the phase of θ at will. Two conventions are often used: $\text{Arg } \theta = \pi/2$ and $\text{Arg } \theta = 0$. We will use the latter. For a % 50 -% 50 beam splitter with $|\theta| = \pi/4$ these leads to the simple transformation law

$$U_{\text{BS}} a U_{\text{BS}}^\dagger = \frac{1}{\sqrt{2}}(a + b) \quad (3.85)$$

$$U_{\text{BS}} b U_{\text{BS}}^\dagger = \frac{1}{\sqrt{2}}(b - a) . \quad (3.86)$$

We remark that this transformation law corresponds to a specific physical realization of a beam splitter which, however, is not realized in practise: imagine the beam splitter to be composed of two prisms with refractive indices $n_>, n_<$ (where $n_> > n_<$) which are glued together. It is well known that light that is reflected from an interface between two dielectric media acquires a phase shift of π if it approaches the interface from the side with the lower refractive index. Imagine that beam a enters the beam splitter through the $n_>$ side. It then does not acquire a phase shift. However, beam b then enters the beam splitter through the $n_<$ medium so that it acquires a phase shift of π when it is reflected and leaves the beam splitter through the a output mode. This π phase factor just corresponds to the minus sign appearing in Eq. (3.86).

3.4.4 Polarization rotator

This is a device that allows to generate any superposition of polarization vectors in a beam of light. It can be realized by a suitable arrangement of half-wave and quarter-wave plates. The latter are pieces of birefringent material that induce a phase shift of π (half-wave) or $\pi/2$ (quarter-wave) between two orthogonal linear polarization states. Here we will follow a very common convention and take horizontal and vertical polarization as basis states. The annihilation operators for photons with horizontal or vertical polarization in beam a are then denoted by a_H and a_V , respectively.

Since a polarization rotator takes a photon with one polarization and transforms it into a photon with another polarization (conserving the number of photons) the phenomenological Hamiltonian for this process should look like

$$H_{\text{PR}} = \zeta a_V^\dagger a_H + \zeta^* a_H^\dagger a_V . \quad (3.87)$$

While the physics is very different, this Hamiltonian is mathematically equivalent to that of a beam splitter. We therefore can immediately write down the action of the corresponding unitary operator

$$U_{\text{PR}} = \exp(\chi a_V^\dagger a_H - \chi^* a_H^\dagger a_V) , \quad (3.88)$$

which leads to the

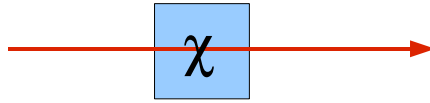
Polarization Rotator rules:

$$U_{PR} a_H U_{PR}^\dagger = a_H \cos |\chi| + a_V \frac{\chi^*}{|\chi|} \sin |\chi| \quad (3.89)$$

$$U_{PR} a_V U_{PR}^\dagger = a_V \cos |\chi| - a_H \frac{\chi}{|\chi|} \sin |\chi| \quad (3.90)$$

$$U_{PR} a_H^\dagger U_{PR}^\dagger = a_H^\dagger \cos |\chi| + a_V^\dagger \frac{\chi}{|\chi|} \sin |\chi| \quad (3.91)$$

$$U_{PR} a_V^\dagger U_{PR}^\dagger = a_V^\dagger \cos |\chi| - a_H^\dagger \frac{\chi^*}{|\chi|} \sin |\chi| \quad (3.92)$$



Graphical representation of the polarization Rotator operation U_{PR}

The half-wave plate itself is actually not a polarization rotator but a phase shifter for a particular linear polarization. If it shifts V relative to H its action is summarized by $\alpha|1_H\rangle + \beta|1_V\rangle \rightarrow \alpha|1_H\rangle - \beta|1_V\rangle$. This can be accomplished by $U_{PS,V}(\pi) = \exp(i\pi a_V^\dagger a_V)$ or $a_V \rightarrow -a_V$. Likewise, a quarter-wave plate corresponds to $U_{PS,V}(\pi) = \exp(i\frac{\pi}{2} a_V^\dagger a_V)$ or $a_V \rightarrow ia_V$.

If the half wave plate is rotated by an angle ϑ it generates a π phase shift for a photon in the state $\cos \vartheta a_V - \sin \vartheta a_H$ and leaves $\cos \vartheta a_H + \sin \vartheta a_V$ invariant. For the mode a_H this behaviour corresponds to the transformation law

$$a_H \rightarrow a_H \cos 2\vartheta + a_V \sin 2\vartheta. \quad (3.93)$$

Similarly, application of a rotated quarter-wave plate then allow us to generate an arbitrary relative phase between the two modes in this supoerposition. In this way U_{PR} can be generated using phase plates.

3.4.5 Polarizing beam splitter

A polarizing beam splitter is a device that transmits horizontal but reflects vertical polarization. It obviously corresponds to a unitary operator that acts on a_V only. We can use the beam splitter operation with $\theta = \pi/2$ to describe this:

Polarizing beam splitter rule:

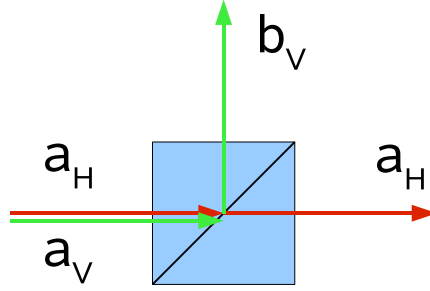
$$U_{PBS} a_H U_{PBS}^\dagger = a_H \quad (3.94)$$

$$U_{PBS} a_V U_{PBS}^\dagger = b_V \quad (3.95)$$

$$U_{PBS} a_H^\dagger U_{PBS}^\dagger = a_H^\dagger \quad (3.96)$$

$$U_{PBS} a_V^\dagger U_{PBS}^\dagger = b_V^\dagger \quad (3.97)$$

It should be mentioned here that an ordinary beam splitter ideally acts on both polarizations in the same way. Thus, if polarization plays a role in an experiment, the beam splitter rules should be applied to the two pairs of differently polarized input modes a_H, b_H and a_V, b_V in the same way, e.g. $a_H \rightarrow (a_H + b_H)/\sqrt{2}$ etc.



Graphical representation of the polarizing beam splitter operation U_{PBS}

3.4.6 Schwinger representation of angular momentum algebra

There is a nice interpretation of linear optical elements in terms of rotations. To see how this works we introduce the Schwinger representation of angular momentum algebra,

$$J_1 = \frac{1}{2}(a^\dagger b + b^\dagger a) \quad (3.98)$$

$$J_2 = \frac{i}{2}(b^\dagger a - a^\dagger b) \quad (3.99)$$

$$J_3 = \frac{1}{2}(a^\dagger a - b^\dagger b) . \quad (3.100)$$

It is easy to check that these operators fulfill the angular momentum algebra $[J_i, J_j] = i\varepsilon_{ijk}J_k$. As usual we can find common eigenstates of \mathbf{J}^2 and J_3 . Some algebra yields

$$\mathbf{J}^2 = \frac{1}{4}(a^\dagger a + b^\dagger b)^2 + \frac{1}{2}(a^\dagger a - b^\dagger b) \quad (3.101)$$

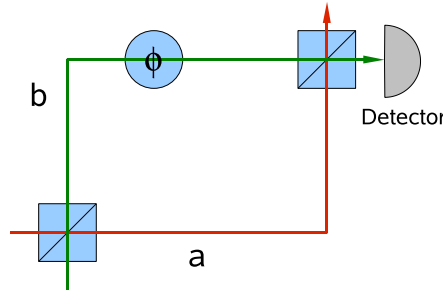
Hence states with a total number N of photons correspond to a representation with $\mathbf{J}^2 = N^2/4 + N/2$. Equating this with the usual angular momentum eigenvalue $l(l+1)$ of \mathbf{J}^2 we find that states with a total number N of photons correspond to a spin $l = N/2$ (irreducible) representation of angular momentum algebra. The magnetic quantum number, i.e., the eigenvalues of J_3 , are just one half of the photon number difference between the two modes.

Angular momentum theory tells us that an operator of the form $U = \exp(2i\zeta \mathbf{n} \cdot \mathbf{J})$ corresponds to a rotation around the axis \mathbf{n} by an angle ζ , where \mathbf{n} is a unit vector. Comparing this to the operators describing linear optical elements above

shows that a beam splitter (and a polarization rotator) just correspond to a rotation by an angle $\zeta = |\theta|$ around the axis $\mathbf{n} = \sin \chi \mathbf{e}_x - \cos \chi \mathbf{e}_y$, where χ is the phase of θ . Furthermore, if we generate a relative phase shift between two modes a and b by applying opposite phase shifters to each mode, $U = U_{\text{PS},a}(\phi)U_{\text{PS},b}(-\phi)$, the result corresponds to a rotation around the z-axis.

3.4.7 Application: Mach-Zehnder interferometer

As a simple application of the linear optics formalism we discuss a Mach-Zehnder experiment as shown in the following figure.



Mach-Zehnder interferometer

It consists of a single photon that enters in mode a (red beam). It passes a %50-%50 beam splitter and acquires a phase shift if it passes through path b (green beam). After passing a second %50-%50 beam splitter we detect whether the photon went through output port b of the second beam splitter.

Mathematically, the input state of the Mach-Zehnder interferometer is given by $|\psi_{\text{in}}\rangle = a^\dagger|0\rangle$. After beam splitter one it is changed to

$$|\psi_1\rangle = U_{\text{BS}}(\pi/4) |\psi_{\text{in}}\rangle = \frac{1}{\sqrt{2}}(a^\dagger + b^\dagger)|0\rangle. \quad (3.102)$$

The application of the phase shifter yields

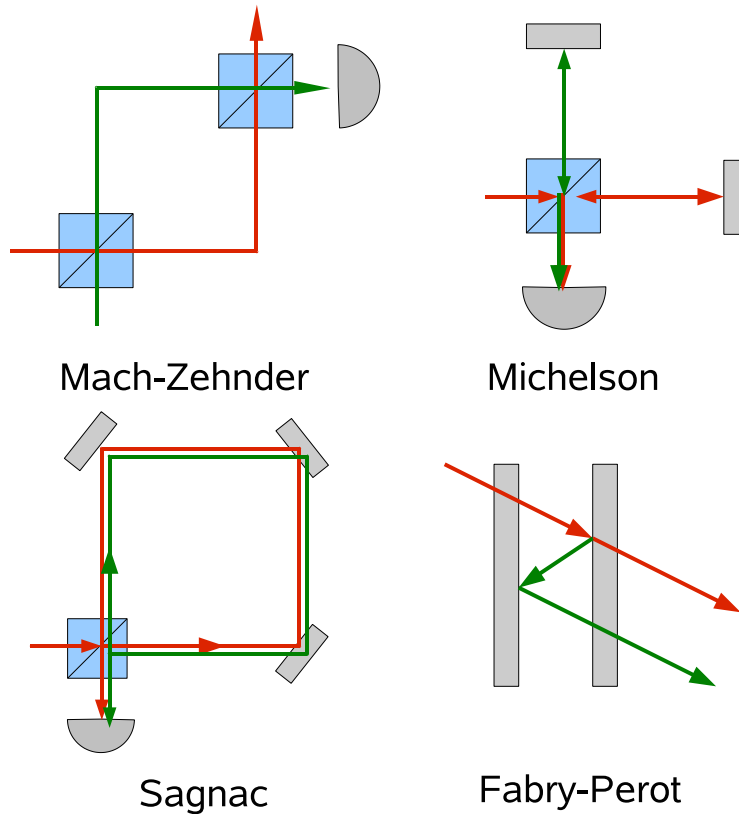
$$|\psi_2\rangle = U_{\text{PS},b}|\psi_1\rangle = \frac{1}{\sqrt{2}}(a^\dagger + b^\dagger e^{i\phi})|0\rangle. \quad (3.103)$$

Finally, the second beam splitter transforms the state to

$$\begin{aligned} |\psi_{\text{out}}\rangle &= U_{\text{BS}}(\pi/4) |\psi_2\rangle \\ &= \frac{1}{2} ((a^\dagger + b^\dagger) + e^{i\phi}(b^\dagger - a^\dagger)) |0\rangle \\ &= \left(a^\dagger \frac{1}{2}(1 - e^{i\phi}) + b^\dagger \frac{1}{2}(1 + e^{i\phi}) \right) |0\rangle \end{aligned} \quad (3.104)$$

The probability to detect the photon in mode b is then given by $P_b = \frac{1}{4}|1 + e^{i\phi}|^2 = \frac{1}{2}(1 + \cos \phi)$. It obviously displays the phenomenon of interference between the two paths of the interferometer because it depends on the relative phase between the two paths.

As a side remark we mention that this calculation can also be applied to different types of interferometers. It only requires that the photon has the possibility to travel along two different paths, but nowhere have we made use of the specific form of these paths. We therefore can use it in the same way to describe a Michelson, Sagnac, or Fabry-Perot interferometer (see figure below). Of course the calculations would differ in a more detailed description which takes into account the evolution of the spatial mode of the photon.



Different types of interferometer

3.5 The Hong-Ou-Mandel dip and non-orthogonal modes

The Hong-Ou-Mandel dip (HOM dip) [48] is a simple and famous demonstration that a naive application of the single harmonic oscillator model is not always appropriate. The basic idea of the HOM dip is that the output state of two photons that simultaneously enter a beam splitter depends strongly on whether they are in the same mode or not. Consider an input state with two photons in different arms of the beam splitter, but in one case they have the same polarization, $|\psi_{\text{in},S}\rangle = a_H^\dagger b_H^\dagger |0\rangle$, whereas in the second case they have different polarization, $|\psi_{\text{in},D}\rangle = a_H^\dagger b_V^\dagger |0\rangle$. The corresponding output states after the (lossless) beam splitter are given by

$$\begin{aligned} |\psi_{\text{out},S}\rangle &= U_{\text{BS}} |\psi_{\text{in},S}\rangle \\ &= \frac{1}{2} (a_H^\dagger + b_H^\dagger) (b_H^\dagger - a_H^\dagger) |0\rangle \\ &= \frac{1}{2} ((b_H^\dagger)^2 - (a_H^\dagger)^2) |0\rangle \end{aligned} \quad (3.105)$$

$$\begin{aligned} |\psi_{\text{out},D}\rangle &= U_{\text{BS}} |\psi_{\text{in},D}\rangle \\ &= \frac{1}{2} (a_H^\dagger + b_H^\dagger) (b_V^\dagger - a_V^\dagger) |0\rangle . \end{aligned} \quad (3.106)$$

This means that if the photons are in exactly the same mode they will interfere in such a way that they always exit the beam splitter together in the same output arm. The probability to find one photon in each of the output arms is zero. On the other hand, if the photons are in orthogonal modes they exit the beam splitter randomly in the same or in different arms.

However, in reality the photons will never be exactly in the same mode, and if one has less than perfect control about when each photon enters the beam splitter their wavepackets might only partially overlap. The input state is therefore a random mix of two photons in modes that are generally non-orthogonal. Therefore, the probability to find one photon in each of the output arms (so-called coincidence measurements) will generally vary with the delay between the photons, which can be controlled by varying the length of the path of one photon before it enters the beam splitter. One therefore will see a dip in the coincidence probability as a function of the delay.

The rest of this Section assumes that you are already familiar with the concept of a density matrix. If this is not the case you may first want to read the first pages of Chapter 7. We assume that the initial state of the photons is given by a mixture

of non-orthogonal Gaussian modes, $\varrho_{\text{in}} = \varrho_{\text{a,in}} \otimes \varrho_{\text{b,in}}$ with

$$\begin{aligned}\varrho_{\text{a,in}} &= \int d\tau f_a(\tau) |1_{a,\phi(\tau)}\rangle \langle 1_{a,\phi(\tau)}| \\ \varrho_{\text{B,in}} &= \int d\tau' f_B(\tau') |1_{B,\psi(\tau')}\rangle \langle 1_{B,\psi(\tau')}| ,\end{aligned}\quad (3.107)$$

with $\int d\tau f_a(\tau) = \int d\tau' f_b(\tau') = 1$. The action of the beam splitter on one of the states $|1_{a,\phi(\tau)}, 1_{b,\psi(\tau')}\rangle$ results in ⁸

$$\begin{aligned}U_{\text{BS}}|1_{a,\phi}, 1_{b,\psi}\rangle &= |\Psi_{1-1}\rangle + |\Psi_{2-0}\rangle \\ &= \frac{1}{2} \left(a_\phi^\dagger b_\psi^\dagger - a_\psi^\dagger b_\phi^\dagger \right) |0\rangle \\ &\quad + \frac{1}{2} \left(b_\phi^\dagger b_\psi^\dagger - a_\psi^\dagger a_\phi^\dagger \right) |0\rangle\end{aligned}\quad (3.108)$$

The probability to detect one photon in each detector is given by

$$\begin{aligned}P_{1-1}(a : \phi(\tau); b : \psi(\tau')) &= \langle \Psi_{1-1} | \Psi_{1-1} \rangle = \\ &= \frac{1}{2} + \frac{1}{4} \left(2 - [a_\phi, a_\psi^\dagger] [b_\psi, b_\phi^\dagger] \right. \\ &\quad \left. - [a_\psi, a_\phi^\dagger] [b_\phi, b_\psi^\dagger] \right)\end{aligned}$$

In the derivation we have used that wavepackets traveling in different beams A,B have zero overlap. The commutators have been calculated in Sec. 3.1.

We thus have shown that for an arbitrary pair of states in the two beams the probability to detect one photon at each output port is given by

$$P_{1-1}(a : \phi(\tau); b : \psi(\tau')) = \frac{1}{2} \left(1 - |\langle \phi(\tau) | \psi(\tau') \rangle|^2 \right) , \quad (3.109)$$

where the scalar product is given by Eq. (3.9) and the modes are normalized positive-frequency modes of the electromagnetic field. Of course one has $P_{2-0}(a : \phi(\tau); b : \psi(\tau')) = 1 - P_{1-1}(a : \phi(\tau); b : \psi(\tau'))$ by conservation of probability (can also be derived explicitly). The total probability to detect a 1-1 output in state ϱ_{in} is then given by

$$\begin{aligned}P_{1-1} &= \int d\tau d\tau' f_a(\tau) f_b(\tau') P_{1-1}(a : \phi(\tau); b : \psi(\tau')) \\ &= \frac{1}{2} - \frac{1}{2} \int d\tau d\tau' f_a(\tau) f_b(\tau') |\langle \phi(\tau) | \psi(\tau') \rangle|^2\end{aligned}\quad (3.110)$$

⁸To keep the notation short we omit here the dependence on τ and τ' .

If the integral on the right-hand side is negligible, we have recovered the result of the simple HOM dip explanation presented at the beginning of this section.

To show that this is the case for realistic pulses we consider the special case of Gaussian wavepackets of the form

$$\phi(\tau) = \sqrt{\frac{\hbar}{2ck\varepsilon_0 cT\sqrt{\pi}}} e^{-\frac{(x-c\tau)^2}{2c^2T^2}} e^{ik(x-c\tau)} \epsilon_\phi \quad (3.111)$$

$$\psi(\tau) = \sqrt{\frac{\hbar}{2ck\varepsilon_0 cT\sqrt{\pi}}} e^{-\frac{(x-c\tau)^2}{2c^2T^2}} e^{ik(x-c\tau)} \epsilon_\psi . \quad (3.112)$$

Here T represents the duration of the pulse, in Alex's experiment about 170 fs. These wavepackets are only different in their polarization vectors ϵ_ϕ and ϵ_ψ , which determine the qubit state of the photons and at the same time determine the scalar product for perfect mode matching,

$$\langle\phi|\psi\rangle_0 \equiv \langle\phi(\tau)|\psi(\tau)\rangle = \epsilon_\phi^* \cdot \epsilon_\psi . \quad (3.113)$$

It is not hard to derive the mode overlap for different times,

$$|\langle\phi(\tau)|\psi(\tau')\rangle|^2 = \frac{4\omega^2 T^4 + (\tau - \tau')^2}{4\omega^2 T^4} e^{-\frac{(\tau - \tau')^2}{2T^2}} |\langle\phi|\psi\rangle_0|^2 \quad (3.114)$$

with $\omega \equiv ck$. If we assume that the mode mismatch probability is given by a Gaussian of width $\Delta\tau$,

$$f_a(\tau) = f_b(\tau) = \frac{e^{-\frac{\tau^2}{\Delta\tau^2}}}{\sqrt{\pi}\Delta\tau} \quad (3.115)$$

we find

$$P_{1-1} = \frac{1}{2} - \frac{1}{2} |\langle\phi|\psi\rangle_0|^2 \frac{\Delta\tau^2 + 4T^2(T^2 + \Delta\tau^2)\omega^2}{4T(T^2 + \Delta\tau^2)^{\frac{3}{2}}\omega^2} \quad (3.116)$$

In the limit that T and $\Delta\tau$ are much larger than $1/\omega$ this becomes

$$P_{1-1} = \frac{1}{2} - \frac{1}{2} |\langle\phi|\psi\rangle_0|^2 \frac{T}{\sqrt{T^2 + \Delta\tau^2}} \quad (3.117)$$

This means we are allowed to use the simple HOM analysis of the previous section whenever the width $\Delta\tau$ of the mode mismatch distribution is much larger than the pulse duration. If the mode mismatch distribution is narrow, we recover the ideal case (i.e., $P_{1-1} = 0$ for identical polarization states).

We can relate this probability to the HOM dip as follows. The HOM dip is a decrease from a constant coincidence (1-1) rate for non-overlapping states to a

smaller (zero, if the states are completely overlapping) coincidence rate if they are pretty much overlapping. If we denote the ratio between these rates as d , then we have

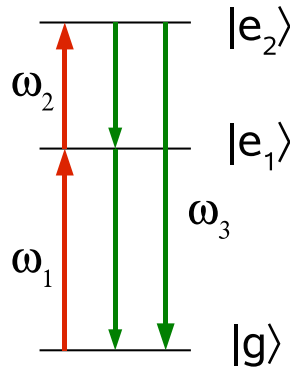
$$\begin{aligned} d &= \frac{P_{1-1}}{P_{1-1}|_{\Delta\tau \rightarrow \infty}} \\ &= 1 - |\langle\phi|\psi\rangle_0|^2 Q \end{aligned} \quad (3.118)$$

with $Q \equiv T/\sqrt{T^2 + \Delta\tau^2}$. This allows us to infer $Q = 1 - d$ where d corresponds to the HOM dip for identical polarization states. Assuming that d does not depend on the polarization we therefore find

$$P_{1-1} = \frac{1}{2} - \frac{1}{2} |\langle\phi|\psi\rangle_0|^2 (1 - d) \quad (3.119)$$

3.6 Aspects of nonlinear optical elements

Nonlinear optical elements, such as a nonlinear crystal like BBO, differ substantially from linear elements and are generally much harder to study theoretically. The polarization $\mathbf{P}(\mathbf{x})$ of a nonlinear medium depends nonlinearly on the electric field of a light beam and can, to some degree, be expanded in a Taylor series of \mathbf{E} . Alternatively, one may assume that the nonlinear dielectric medium generates an effective interaction between photons. A simple example of a physical process in which such an interaction is generated is the case of a three-level atom in cascade configuration (see figure; for an introduction into few-level models for atoms see chapter 6). In absence of light the atoms are in the lowest energy level $|g\rangle$. A photon of frequency ω_1 can pump the atoms into the first excited state $|e_1\rangle$. Only if this process took place can a photon of frequency ω_2 pump the atoms from $|e_1\rangle$ to $|e_2\rangle$. Obviously the optical properties for photon 2 depend on the presence of photon 1, i.e., an effective interaction between these photons is generated.



A simple cascade model for atoms in a nonlinear medium.

This interaction may result in different effects. For instance, the atom may decay through the same channel through which it has been excited, thus creating photons of the same frequency which it had previously absorbed. Denoting the annihilation operator for a photon of frequency ω_i by a_i we then can expect to have an effective interaction Hamiltonian of the form $H_{\text{int}} = \kappa a_1^\dagger a_2^\dagger a_2 a_1$, where κ is a coupling constant that determines the strength of the effective interaction. Of particular interest is the case when $\omega_1 = \omega_2$ so that these two photon modes are equal, $a_1 = a_2 = a$. We then find a Kerr-type nonlinearity with

$$H_{\text{Kerr}} = \kappa (a^\dagger)^2 a^2 \quad (3.120)$$

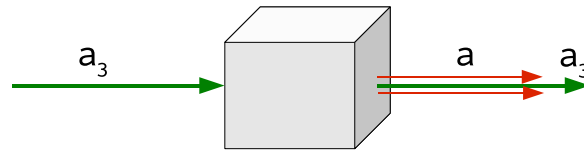
which is a very common model in quantum optics. Another important process is up- and down-conversion. If the atom decays through mode ⁹ a_3 two photons are absorbed and one other is created. The corresponding effective interaction Hamiltonian would look like $H_{\text{int}} = \kappa a_3^\dagger a_2 a_1 + \kappa^* a_3 a_2^\dagger a_1^\dagger$. The first part describes up-conversion in which two photons of lower energy are converted into one photon of higher energy. Energy conservation implies that $\omega_3 = \omega_1 + \omega_2$ must hold. The second part describes down-conversion whereby two lower-energy photons are created from one photon of higher energy. For $\omega_1 = \omega_2$ one finds

$$H_{\text{conv}} = \kappa a_3^\dagger a^2 + \kappa^* a_3 (a^\dagger)^2 \quad (3.121)$$

which is a very important model for the creation of entangled photons and for squeezing.

3.6.1 Squeezed states of light

Let us consider the situation in the figure below: a nonlinear crystal whose effect can be approximated by H_{conv} is subject to an intense classical field of frequency ω_3 .



Basic principle of a squeezing experiment

If the change in the input beam can be neglected we can replace the operator a_3 by the coherent state amplitude α_3 of the classical field so that we get an effective Hamiltonian for mode a alone,

$$H_{\text{sq}} = g a^2 + g^* (a^\dagger)^2, \quad (3.122)$$

⁹Selection rules exclude this possibility for atoms in free space. However, the process is possible in a crystal or if external fields are applied.

In the same way as for linear optical elements we can deduce that the unitary squeezing operator, which is usually denoted as $S(\beta)$, is given by

$$S(\beta) = \exp(\beta(a^\dagger)^2 - \beta^* a^2), \quad (3.123)$$

Again we can use Eq. (3.72) to derive the transformation properties for the annihilation and creation operator. As in the case of the beam splitter the series can be separated into even and odd l and the corresponding sums can be identified with the Taylor expansion of cosh and sinh. This leads to the

Squeezing rule:

$$SaS^\dagger = a \cosh(2|\beta|) - a^\dagger \frac{\beta}{|\beta|} \sinh(2|\beta|) \quad (3.124)$$

$$Sa^\dagger S^\dagger = a^\dagger \cosh(2|\beta|) - a \frac{\beta^*}{|\beta|} \sinh(2|\beta|) \quad (3.125)$$

This transformation law, in which annihilation and creation operators are mixed, is very famous and also called a Bogoliubov transformation. It appears frequently in theoretical physics when a system is governed by a nonlinear Hamiltonian and one linearizes the dynamical equations for small perturbations around a given mean field solution. An example would be the description of quasi-particles in a Bose-Einstein condensate. In our case, the mean field solution is given by the intense pump field and the small perturbations are given by photons in mode a . Another, more exotic application of Bogoliubov transformations which does not rely on linearization is particle creation in a curved space time. For instance, Hawking radiation can also be described using a Bogoliubov transformation.

Before we explore the features of squeezed states of light we remark that squeezing is not restricted to a single mode. In general the pump field may generate two photons in different modes a and b . The interaction Hamiltonian is then replaced by $H_{sq2} = gab + g^* a^\dagger b^\dagger$. It is not hard to see that this leads to the

Two-mode squeezing rule:

$$U_{sq2} a U_{sq2}^\dagger = a \cosh(2|\beta|) - b^\dagger \frac{\beta}{|\beta|} \sinh(2|\beta|) \quad (3.126)$$

$$U_{sq2} a^\dagger U_{sq2}^\dagger = a^\dagger \cosh(2|\beta|) - b \frac{\beta^*}{|\beta|} \sinh(2|\beta|) \quad (3.127)$$

$$U_{sq2} b U_{sq2}^\dagger = b \cosh(2|\beta|) - a^\dagger \frac{\beta}{|\beta|} \sinh(2|\beta|) \quad (3.128)$$

$$U_{sq2} b^\dagger U_{sq2}^\dagger = b^\dagger \cosh(2|\beta|) - a \frac{\beta^*}{|\beta|} \sinh(2|\beta|) \quad (3.129)$$

Let us now explore the physical content of the single-mode squeezing transformation (3.125). We start with calculating the mean number of particles in a squeezed vacuum state $|0_{\text{sq}}\rangle = S(\beta)|0\rangle$. Using the fact that $S^\dagger(\beta) = S(-\beta)$ one finds

$$\begin{aligned}
 \bar{n} &= \langle 0_{\text{sq}} | a^\dagger a | 0_{\text{sq}} \rangle \\
 &= \langle 0 | S^\dagger a^\dagger a S | 0 \rangle \\
 &= \langle 0 | (S^\dagger a^\dagger S) (S^\dagger a S) | 0 \rangle \\
 &= \langle 0 | \left(a^\dagger \cosh(2|\beta|) + a \frac{\beta^*}{|\beta|} \sinh(2|\beta|) \right) \left(a \cosh(2|\beta|) + a^\dagger \frac{\beta}{|\beta|} \sinh(2|\beta|) \right) | 0 \rangle \\
 &= \langle 0 | a \frac{\beta^*}{|\beta|} \sinh(2|\beta|) a^\dagger \frac{\beta}{|\beta|} \sinh(2|\beta|) | 0 \rangle \\
 &= \sinh^2(2|\beta|)
 \end{aligned} \tag{3.130}$$

Hence a squeezed vacuum has a non-zero mean number of particles. The physical reason is simple: down conversion in the nonlinear crystal populates the initially empty mode a by creating pairs of photons. The energy for this process is provided by the pump beam. This is an example of an active optical element. An especially interesting feature of a squeezed vacuum becomes obvious in the number representation,

$$|0_{\text{sq}}\rangle \sim |0\rangle + 2|\beta||2\rangle + \dots, \tag{3.131}$$

i.e., a squeezed vacuum contains only even number states. In particular, a weakly squeezed vacuum state can be considered as a superposition of a two-photon state with the vacuum which can be used to create entangled two-photon states.

However, the great interest in squeezed states is not related to photon production but to the fact that the mean deviation for certain observables can be suppressed below their vacuum value. To shed light on this consider the case that mode a is initially in a coherent state $|\alpha\rangle$. Down-conversion then transforms this state into a squeezed coherent state $|\alpha_{\text{sq}}\rangle = U_{\text{sq}}|\alpha\rangle = U_{\text{sq}}D(\alpha)|0\rangle$. To experimentally analyze its properties one generally use the quadrature operators

$$\begin{aligned}
 X(\theta) &= \frac{1}{\sqrt{2}}(a + a^\dagger) \cos \theta + \frac{i}{\sqrt{2}}(a^\dagger - a) \sin \theta \\
 &= \frac{1}{\sqrt{2}}(ae^{-i\theta} + a^\dagger e^{i\theta})
 \end{aligned} \tag{3.132}$$

$$\begin{aligned}
 Y(\theta) &= \frac{i}{\sqrt{2}}(a^\dagger - a) \cos \theta - \frac{1}{\sqrt{2}}(a + a^\dagger) \sin \theta \\
 &= \frac{i}{\sqrt{2}}(a^\dagger e^{i\theta} - ae^{-i\theta})
 \end{aligned} \tag{3.133}$$

For $\theta = 0$ these operators coincide with the position and momentum operator of a harmonic oscillator. For each choice of θ they form a pair of conjugate observables because $[X(\theta), Y(\theta)] = i$. We have seen in our discussion of coherent states that for $\theta = 0$ they are proportional to the electric and magnetic field operator in specific situations. In general situations X and Y are proportional to E and B for some value of θ , but they also test other features of the quantum state of light as we will now see.

The mean value of X and Y for a squeezed coherent state are given by

$$\begin{aligned}\langle X \rangle &= \langle 0 | D^\dagger S^\dagger X S D | 0 \rangle \\ &= \langle 0 | D^\dagger \frac{1}{\sqrt{2}} (a e^{-i\theta} + a^\dagger e^{i\theta}) D | 0 \rangle \\ &= \langle 0 | \frac{1}{\sqrt{2}} ((a + \alpha) e^{-i\theta} + (a^\dagger + \alpha^*) e^{i\theta}) | 0 \rangle \\ &= \frac{1}{\sqrt{2}} (\alpha e^{-i\theta} + \alpha^* e^{i\theta})\end{aligned}\tag{3.134}$$

$$\langle Y \rangle = \frac{i}{\sqrt{2}} (\alpha^* e^{i\theta} - \alpha e^{-i\theta})\tag{3.135}$$

The calculation of the variances follows the same principles but is tedious and left to the reader as an exercise. The result is

$$\Delta X^2 = \frac{1}{2} (\text{ch}^2 + \text{sh}^2 + 2\text{ch sh} \cos(2\theta - \varphi))\tag{3.136}$$

$$\Delta Y^2 = \frac{1}{2} (\text{ch}^2 + \text{sh}^2 - 2\text{ch sh} \cos(2\theta - \varphi))\tag{3.137}$$

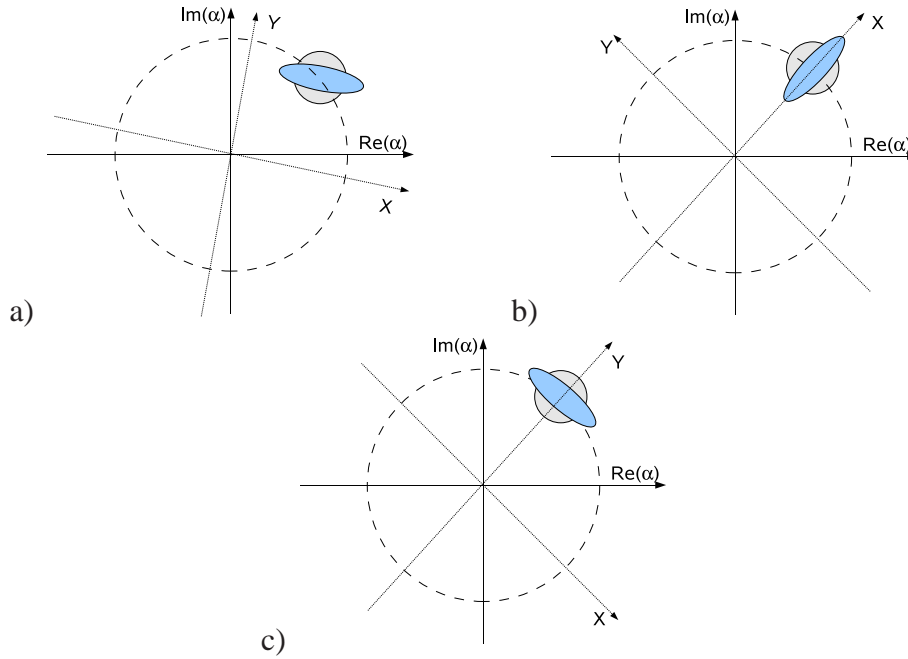
with $\beta/|\beta| = \exp(i\varphi)$, $\text{ch} = \cosh(2|\beta|)$ and $\text{sh} = \sinh(2|\beta|)$. For the special choice quadrature operators that corresponds to $\theta = \varphi/2$ we find

$$\Delta X = \frac{1}{\sqrt{2}} e^{2|\beta|}\tag{3.138}$$

$$\Delta Y = \frac{1}{\sqrt{2}} e^{-2|\beta|}\tag{3.139}$$

Hence the noise in Y is reduced as compared to the shot noise (i.e., the noise of Y in the vacuum state) while the noise of X is increased. Note that for this case we have $\Delta X \Delta Y = 1/2$ which indicates that squeezed states are states of minimum uncertainty, i.e., they fulfill Heisenberg's uncertainty relation for X and Y with an equality sign.

This feature is very useful for a variety of reasons. To see this we represent a squeezed coherent state graphically in a similar way as the ordinary coherent state:

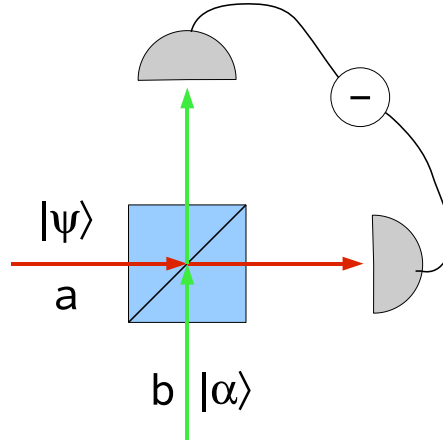


Graphical representation of squeezed coherent states.

Examples a,b,c represent three different squeezed coherent states of light. Each state is represented by an ellipse whose center is at $\langle X \rangle + i\langle Y \rangle$ with respect to the X, Y coordinate system. The corresponding unsqueezed coherent state is represented by a (grey) circle whose area indicates the uncertainty of a quadrature measurement. A squeezed coherent state. The ellipse of the squeezed coherent states indicates that the noise for X is increased and for Y is reduced. Example a) corresponds to a general squeezed coherent state. If the ellipse is arranged as in example b) we have reduced noise when measuring the phase of the complex amplitude and increased noise when measuring in modulus. This is of practical importance in interferometry where improved knowledge of the phase (at the cost of increased fluctuations in the photon number) is desirable. Example c) would allow to have improved knowledge of the number of photons in mode a at the cost of increasing the noise in the phase. In a sense one can consider a photon number state $|n\rangle$ as the limit of infinite squeezing of the photon number.

Homodyne detection

The question remains how to measure the quadrature operators $X(\theta)$ and $Y(\theta)$. The standard technique to accomplish this is homodyne detection.



Homodyne detection to measure quadrature operators.

We want to measure the mean value of the quadrature operators for a quantum state $|\psi\rangle$ of a signal light beam in mode a . To do so we use a %50-%50 beam splitter to combine this light field with a classical light beam in mode b that is called local oscillator and is described by a coherent state $|\alpha\rangle$. This field is much more intense than the signal beam, i.e., the mean number $|\alpha|^2$ of photons in mode b is much larger than $\langle\psi|a^\dagger a|\psi\rangle$. The resulting input state can be written as $|\psi_{\text{in}}\rangle = D_b(\alpha)f(a^\dagger)|0\rangle$ where we have represented the state $|\psi\rangle$ of mode a in the form $|\psi\rangle = f(a^\dagger)|0\rangle$ with f some Taylor-expandable function of the creation operator. In practise, f is a polynomial of low order. $D_b(\alpha)$ denotes the shift operator for mode b .

We measure the photo current at each output channel of the beam splitter and subtract the results. Because the photo current is approximately proportional to the mean photon number measured by the detector the output signal ΔI is proportional to

$$\begin{aligned}
 \Delta I &= \langle\psi_{\text{in}}|U_{\text{BS}}^\dagger(a^\dagger a - b^\dagger b)U_{\text{BS}}|\psi_{\text{in}}\rangle \\
 &= \langle\psi_{\text{in}}|\frac{1}{2}((a^\dagger - b^\dagger)(a - b) - (b^\dagger + a^\dagger)(b + a))|\psi_{\text{in}}\rangle \\
 &= -\langle\psi_{\text{in}}|(a^\dagger b + b^\dagger a)|\psi_{\text{in}}\rangle \\
 &= -\langle 0|f^\dagger D_b^\dagger(a^\dagger b + b^\dagger a)D_b f|0\rangle \\
 &= -\langle 0|f^\dagger(a^\dagger(b + \alpha) + (b^\dagger + \alpha^*)a)f|0\rangle \\
 &= -\langle\psi|(a^\dagger \alpha + \alpha^* a)|\psi\rangle
 \end{aligned} \tag{3.140}$$

Hence, if the phase θ of the local oscillator ($\alpha = |\alpha|e^{i\theta}$) is fixed, ΔI is proportional to $\langle X(\theta)\rangle$.

3.6.2 Nonlinear media of Kerr type

Together with parametric up- and down-conversion, the Kerr effect is one of the most important nonlinear effects in quantum optics. In classical electrodynamics the Kerr effect can simply be described as an intensity-dependent index of refraction,

$$n = n_0 + n_2 I, \quad (3.141)$$

where I is the intensity of the field and n_2 the non-linear refractive index. Typically n_2 is very small so that large intensities are needed to create a significant effect; in (fused silica) optical fibres, for instance, one has $n_2 \approx 3 \times 10^{-16} \text{ cm}^2/\text{W}$. However, in media exhibiting electromagnetically induced transparency n_2 can be as large as $0.1 \text{ cm}^2/\text{W}$ [21], although in this case there are practical restrictions on the intensity of the beam. Because a plane electromagnetic wave of frequency ω varies in a dielectric as $\exp(inkz - \omega t)$, where $k = \omega/c$ is the wavenumber in vacuum, we see that a variation of the intensity I changes the phase of the wave as $\exp(in_2 I k z)$. If this effect happens with a single light beam it is called self phase modulation. In some media the intensity I_2 of a second beam (with different polarization and/or frequency) changes the phase of the first beam. In this case the refractive index n_1 of the first beam varies as $n_1 = n_0 + n_2 I_2$ and one speaks of cross phase modulation.

To construct a quantum-version of the Kerr effect we start by considering the intensity operator. The classical intensity of a beam of light can be derived from the Poynting vector

$$\mathbf{S} = \frac{1}{\mu_0} \mathbf{E} \times \mathbf{B} \quad (3.142)$$

This vector describes the change of energy of the electromagnetic field inside some volume. The energy density is given by

$$u = \frac{\varepsilon_0}{2} \mathbf{E}^2 + \frac{1}{2\mu_0} \mathbf{B}^2 \quad (3.143)$$

so that, with V an arbitrary volume in free space and ∂V its boundary,

$$\begin{aligned}
\frac{dE}{dt} &= \int_V d^3x \frac{du}{dt} \\
&= \int_V d^3x \left(\frac{1}{\mu_0} \mathbf{B} \cdot \dot{\mathbf{B}} + \varepsilon_0 \mathbf{E} \cdot \dot{\mathbf{E}} \right) \\
&= \frac{1}{\mu_0} \int_V d^3x (-\mathbf{B} \cdot \nabla \times \mathbf{E} + \mathbf{E} \cdot \nabla \times \mathbf{B}) \\
&= -\frac{1}{\mu_0} \int_V d^3x \nabla \cdot (\mathbf{E} \times \mathbf{B}) \\
&= - \int_V d^3x \nabla S \\
&= - \oint_{\partial V} d^2\mathbf{A} \cdot \mathbf{S}
\end{aligned} \tag{3.144}$$

where $d^2\mathbf{A}$ is the surface element on the boundary ∂V which is a vector whose direction is normal to the surface. In the derivation we have used Gauss' law and Maxwell's equations in free space.

An operator for the field intensity can be found by replacing the fields in the Poynting vector by the respective operators. However, there are some subtleties that have to be addressed. The complete operator would look like

$$\hat{S} \stackrel{?}{=} \frac{1}{\mu_0} (\hat{\mathbf{E}}^{(+)} \times \hat{\mathbf{B}}^{(+)} + \hat{\mathbf{E}}^{(-)} \times \hat{\mathbf{B}}^{(+)} + \hat{\mathbf{E}}^{(+)} \times \hat{\mathbf{B}}^{(-)} + \hat{\mathbf{E}}^{(-)} \times \hat{\mathbf{B}}^{(-)}) . \tag{3.145}$$

The first and the last term are very rapidly oscillating (as $\exp(\pm 2i\omega t)$) and therefore would average out over the time scale on which a detector reacts. They are therefore dropped. The third term would give a contribution even when the state is the vacuum, which is undesirable. For this reason, and as it is often done in quantum field theories to avoid non-zero vacuum expectation values, one exchanges the sequence of the operators so that all annihilation operators are to the right of all creation operators. This is called normal ordering. We therefore find for the Poynting operator the expression¹⁰

$$\hat{S} = \frac{1}{\mu_0} (\hat{\mathbf{E}}^{(-)} \times \hat{\mathbf{B}}^{(+)} - \hat{\mathbf{B}}^{(-)} \times \hat{\mathbf{E}}^{(+)}) . \tag{3.146}$$

In most cases in quantum optics one is interested in the behaviour of a well focused beam or pulse of light. This was the fundamental assumption behind the paraxial approximation, for instance. If all photons in the system are composed

¹⁰The minus sign stems from rewriting the normally ordered form in term of the vector product of B and E .

of superpositions of plane waves which are close to a central wavevector $\mathbf{k} = k\mathbf{n}$, where \mathbf{n} is a unit vector, then we can use the plane-wave relation $\mathbf{B} = (\mathbf{k} \times \mathbf{E})/\omega_k = \mathbf{n} \times \mathbf{E}/c$ along with $\mathbf{E} \cdot \mathbf{k} \approx 0$. This reduces the Poynting operator to

$$\hat{\mathbf{S}} \approx 2c\varepsilon_0 \hat{\mathbf{E}}^{(-)} \cdot \hat{\mathbf{E}}^{(+)} \mathbf{n} . \quad (3.147)$$

This suggests to define the intensity operator, which describes the power of the pulse flowing through a unit area perpendicular to the radiation direction, as

$$\hat{I}(\mathbf{x}) = 2c\varepsilon_0 \hat{\mathbf{E}}^{(-)}(\mathbf{x}) \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{x}) . \quad (3.148)$$

Let us now develop an intuition of what we would expect for a single mode theory of a Kerr medium. When classical light propagates along, say, the z -axis, it generally acquires a phase factor of the form $\exp(iknz)$. The nonlinear phase shift is therefore given by $\exp(ikn_2Iz)$, where z is the length of the nonlinear medium. Ignoring the difference between group and phase velocity for this estimation, the time the light travels through the medium is given by $t = z/c' = zn_0/c$, where c' is the speed of light in the medium. Hence, the nonlinear phase shift can be written as $\exp(ikn_2Ict/n_0)$.

Turning to quantum mechanics we replace the intensity by the corresponding operator. In single-mode approximation the electric field operator becomes $\hat{\mathbf{E}} \approx \mathbf{E}_a(\mathbf{x})a$, where $\mathbf{E}_a(\mathbf{x})$ is the mode function associated with a . The intensity operator therefore becomes

$$\hat{I}(\mathbf{x}) \approx 2c\varepsilon_0 |\mathbf{E}_a(\mathbf{x})|^2 a^\dagger a . \quad (3.149)$$

Since a classical field amplitude acquires a phase shift $\exp(ikn_2Ict/n_0)$, the corresponding quantum mechanical observable should be modified by the operator $\exp(ikn_2\hat{I}ct/n_0)$. From our discussion of coherent states we know that this observable is the annihilation operator a . We therefore expect a phase change of the quantized radiation that is proportional to $a^\dagger a$. The Kerr Hamiltonian

$$H_{\text{SPM}} = \frac{\hbar\kappa}{2} (a^\dagger)^2 a^2 \quad (3.150)$$

accomplishes this for self phase modulation (SPM). For cross phase modulation (XPM) one would have

$$H_{\text{XPM}} = \hbar\kappa a^\dagger b^\dagger ba . \quad (3.151)$$

For the case of self phase modulation, let us consider the Heisenberg equation of motion for a ,

$$\begin{aligned} i\hbar\partial_t a &= [a, H_{\text{int}}] \\ &= \kappa a^\dagger a^2 , \end{aligned} \quad (3.152)$$

which has the solution

$$a(t) = e^{-i\kappa t a_0^\dagger a_0} a_0 , \quad (3.153)$$

with $a_0 \equiv a(0)$ the annihilation operator in Schrödinger picture. Such a Hamiltonian can for instance be achieved by a intensity-dependent energy level shift in resonant atoms. We will see how such level shifts are generated when we discuss adiabatic elimination (see Sec. 6.4.2).

We can now compare this solution to the classically derived expression for self phase modulation. In Sec. 3.4.2 we have seen that the phase shift acquired by the annihilation operator is just the negative of the phase shift acquired by the complex amplitude of a coherent state. We therefore can make the identification

$$\kappa = 2kc^2\varepsilon_0 \frac{n_2}{n_0} |\mathbf{E}_a(\mathbf{x})|^2 , \quad (3.154)$$

which obviously depends on the mode. For the Gaussian mode (3.42) the maximum amplitude is given by Eq. (3.44). This leads to a nonlinear coefficient of the form

$$\kappa_G = \omega \frac{n_2}{n_0} \frac{\hbar\omega}{\tau A \sqrt{\pi}} , \quad (3.155)$$

where $A = \pi w_0^2$ is the transverse area of the mode and τ the duration of the pulse. This is easy to interpret. $P_0 = \hbar\omega/\tau$ corresponds to the power needed to emit a single photon during time τ . $I_0 = \hbar\omega/(\tau A)$ is the corresponding intensity of a single-photon pulse if it is confined to a transverse area A . For a $\tau = 130$ fs pulse of a single telecom wavelength photon ($\lambda = 1.3\mu\text{m}$) in an optical fibre with a core diameter of $2w_0 = 5\mu\text{m}$ this amounts to $P_0 = 1\mu\text{W}$, $I_0 = 6\text{ W/cm}^2$, and $\kappa_G = 10^{-15}\omega$. In other words, to achieve a phase shift of $\kappa_G t = 1$ rad, a single photon would need to travel for $z = ct \approx 2 \cdot 10^5$ km through an optical fibre, which clearly is impossible because of losses. To achieve a 1 rad phase shift over a distance of 1 km the pulse peak power would need to be $P = nP_0 = 0.25\text{ W}$, corresponding to a pulse with $2 \cdot 10^5$ photons in it.

Eq. (3.153) allows us to derive the time evolution of a general quantum state of light under the influence of SPM, but it is easier to directly calculate the time evolution using the Hamiltonian. With an expansion in the number state basis, $|\psi(t)\rangle = \sum_n \psi_n(t) |n\rangle$, we get the Schrödinger equation

$$i\partial_t \psi_n = \frac{\kappa}{2} n(n-1) \psi_n , \quad (3.156)$$

so that

$$|\psi(t)\rangle = \sum_n \psi_n(0) e^{-i\kappa t n(n-1)/2} |n\rangle . \quad (3.157)$$

In particular, a coherent state evolves into

$$|\psi(t)\rangle = e^{-|\alpha|^2/2} \sum_n \frac{\alpha^n}{\sqrt{n!}} e^{-i\kappa t n(n-1)/2} |n\rangle, \quad (3.158)$$

which is not a coherent state anymore (because the coefficients are not of the form $\alpha^n(t)/\sqrt{n!}$). Hence, in SPM a classical state evolves into a non-classical state.

Application: controlled phase gate

An important application of XPM would be the optical implementation of a controlled phase-gate for quantum information [49]. Like the controlled not-gate, the c-phase gate together with all possible single-qubit operations would enable universal quantum computation. For the logical qubit states $|0\rangle, |1\rangle$ and the corresponding two-qubit state $|00\rangle, \dots$, the c-phase gate has the following truth table,

$$|i, j\rangle \rightarrow (-1)^{ij} |i, j\rangle, \quad i, j = 0, 1. \quad (3.159)$$

That is, the logical state $|11\rangle$ acquires a π phase shift and all other basis states are unchanged.

In principle it is easy to achieve this using XPM. Let us consider two-photon states where each photon is in a different spatial mode a, b and can have horizontal or vertical polarization. The encoding of a qubit is given by the polarization of each photon, say $0 \leftrightarrow H$ and $1 \leftrightarrow V$. For instance, the logical state $|01\rangle$ then corresponds to the physical state $a_H^\dagger b_V^\dagger |\text{vac}\rangle$, where we denote the vacuum state by $|\text{vac}\rangle$ to avoid confusion with the logical states. We now assume we have a nonlinear crystal that generates XPM only if the photons have vertical polarization. This is described by the Hamiltonian

$$H_{\text{XPM}} = \hbar\kappa a_V^\dagger a_V b_V^\dagger b_V. \quad (3.160)$$

Then the initial state

$$|\psi_{\text{in}}\rangle = \alpha_{00} a_H^\dagger b_H^\dagger |\text{vac}\rangle + \alpha_{01} a_H^\dagger b_V^\dagger |\text{vac}\rangle + \alpha_{10} a_V^\dagger b_H^\dagger |\text{vac}\rangle + \alpha_{11} a_V^\dagger b_V^\dagger |\text{vac}\rangle \quad (3.161)$$

evolves into

$$|\psi_{\text{out}}\rangle = \alpha_{00} a_H^\dagger b_H^\dagger |\text{vac}\rangle + \alpha_{01} a_H^\dagger b_V^\dagger |\text{vac}\rangle + \alpha_{10} a_V^\dagger b_H^\dagger |\text{vac}\rangle + e^{-i\kappa t} \alpha_{11} a_V^\dagger b_V^\dagger |\text{vac}\rangle. \quad (3.162)$$

If the photons propagate through the crystal for a time $t = \pi/\kappa$ we have accomplished an optical c-phase gate.

However, there are several problems with this scheme. The most important one is that for nonlinear crystals the constant κ is so small that $\kappa t \ll 1$ in all practical cases. In recent years the possibility to achieve large XPM nonlinearities in atomic gases (see, e.g., Ref. [50]) has generated new hope in constructing an optical c-phase gate, but as we will see in the next two subsections even this approach may face some severe problems.

3.6.3 Shortcomings of the single-mode approximation I: Mode distortion

While it is appealingly simple for a nonlinear theory, the single-mode approximation for Kerr nonlinearities is only of restricted value. Like in all nonlinear processes, the assumption that all photons occupy a single mode is often inappropriate, even if this mode function is adapted to the nonlinear problem.

Let us evaluate this problem in a simple one-dimensional multi-mode theory, where we replace the positive-frequency part $\mathbf{E}^{(+)}(\mathbf{x})$ of the vector operator of the electric field by a scalar operator $\mathcal{E}(z)$ that fulfills the commutation relation

$$[\mathcal{E}(z), \mathcal{E}^\dagger(z')] = \frac{\hbar\omega_0}{2\varepsilon_0 F} \delta(z - z') \equiv \eta \delta(z - z'), \quad (3.163)$$

This model, which is often called scalar QED, neglects the transverse dispersion and the polarization degree-of-freedom of a light beam, but is otherwise similar to ordinary QED. The area F can be interpreted as the area of the light pulse transverse to the z -axis along which it propagates. For instance, it could be identified with the area of the core of a single-mode optical fibre. $\mathcal{E}(z)$ creates a (scalar) photon at position z . To create a photon in a particular mode, e.g., a Gaussian wave packet $\phi(z)$, we can introduce the corresponding annihilation operator as

$$a_\phi = \frac{1}{\eta} \int dz \phi^*(z) \mathcal{E}(z), \quad (3.164)$$

which ensures that $[\mathcal{E}(z), a_\phi^\dagger] = \phi(z)$ so that we can extract the mode function from the commutator.

We assume that the nonlinear Hamiltonian is of Kerr-type at each point inside the medium. We therefore can write it as

$$H = \frac{\kappa}{2} \int dz' (\mathcal{E}^\dagger(z'))^2 \mathcal{E}^2(z'). \quad (3.165)$$

Let us consider the dynamics of an initial state which is a superposition of a one-photon state and a two-photon state,

$$|\psi(0)\rangle = \frac{1}{\sqrt{2}} a_\phi^\dagger |0\rangle + \frac{1}{2} (a_\phi^\dagger)^2 |0\rangle \quad (3.166)$$

Because the Kerr Hamiltonian does not change the number of photons we can write the state at any time as a superposition of a general one- and two-photon state, i.e.,

$$|\psi(t)\rangle = \int dz P(z, 0) \mathcal{E}^\dagger(z) |0\rangle + \int dz dz' Q(z, z', 0) \mathcal{E}^\dagger(z) \mathcal{E}^\dagger(z') |0\rangle$$

with the time dependent functions $P(z, t)$ and $Q(z, z', t)$ fulfilling the initial conditions

$$P(z, 0) = \frac{1}{\sqrt{2\eta}} \phi(z) \quad (3.167)$$

$$Q(z, z', 0) = \frac{1}{2\eta^2} \phi(z) \phi(z') \quad (3.168)$$

The Schrödinger equation $i\partial_t|\psi\rangle = H|\psi\rangle$ then yields the differential equations

$$i\partial_t P(z, t) = 0 \quad (3.169)$$

$$i\partial_t Q(z, z', t) = \kappa\eta^2 \delta(z - z') Q(z, z', t) \quad (3.170)$$

This means that the single-photon part is unperturbed, $P(z, t) = P(z, 0)$. However, the two-photon part will evolve into a state that cannot be written as a product of two mode functions anymore, $Q(z, z', t) \neq \phi_1(z, t) \phi_2(z', t)$. This means that the two photons become entangled, it then wrong to assume that there is a mode function that is occupied by both photons.

While it would be instructive to go in detail through this calculation, it is somewhat plagued by the appearance of the δ distribution in Eq. (3.170). This is a consequence of our over-simplified Hamiltonian where we tacitly assumed that we have a point-like interaction,

$$H = \frac{\kappa}{2} \int dz dz' \mathcal{E}^\dagger(z') \mathcal{E}^\dagger(z) \delta(z - z') \mathcal{E}(z) \mathcal{E}(z'), \quad (3.171)$$

rather than having an interaction potential of finite range,

$$H = \frac{1}{2} \int dz dz' \mathcal{E}^\dagger(z') \mathcal{E}^\dagger(z) V(z - z') \mathcal{E}(z) \mathcal{E}(z'). \quad (3.172)$$

A treatment based on an extended interaction potential $V(z - z')$ is possible but beyond the scope of this course.

3.6.4 Shortcomings of the single-mode approximation II: classical SPM

Potential problems of the single-mode approximation can also be seen in the evolution of a classical state. Let us check if the Kerr Hamiltonian does correctly reproduce self-phase modulation of a classical beam of light, i.e., let us calculate the evolution of the mean electric field amplitude if the initial state is a coherent state. One easily finds in Heisenberg picture

$$\begin{aligned} E(t) &= \langle \alpha | a(t) | \alpha \rangle \\ &= \langle \alpha | e^{-i\kappa t a_0^\dagger a_0} a_0 | \alpha \rangle \\ &= \alpha \langle \alpha | e^{-i\kappa t a_0^\dagger a_0} | \alpha \rangle. \end{aligned} \quad (3.173)$$

This can be evaluated using

Proposition 1:

$$\langle \alpha | e^{i\kappa t a_0^\dagger a_0} | \alpha \rangle = \exp(|\alpha|^2(e^{i\kappa t} - 1)) . \quad (3.174)$$

Proof: Let $I(t) \equiv \langle \alpha | e^{i\kappa t a_0^\dagger a_0} | \alpha \rangle$. Clearly $I(0) = 1$. The time derivative of I is given by

$$\begin{aligned} \partial_t I &= i\kappa \langle \alpha | a_0^\dagger a_0 e^{i\kappa t a_0^\dagger a_0} | \alpha \rangle \\ &= i\kappa \alpha^* \langle \alpha | a_0 e^{i\kappa t a_0^\dagger a_0} | \alpha \rangle . \end{aligned} \quad (3.175)$$

To move a_0 to the right we note that $a_0 e^{i\kappa t a_0^\dagger a_0} = e^{i\kappa t a_0^\dagger a_0} (e^{-i\kappa t a_0^\dagger a_0} a_0 e^{i\kappa t a_0^\dagger a_0})$. The term inside the parentheses is equivalent to the free time evolution of a harmonic oscillator of frequency $-\kappa$. It is well known that the latter is given by $e^{i\kappa t} a_0$. We therefore find

$$\begin{aligned} \partial_t I &= i\kappa \alpha^* \langle \alpha | e^{i\kappa t a_0^\dagger a_0} e^{i\kappa t} a_0 | \alpha \rangle \\ &= i\kappa |\alpha|^2 e^{i\kappa t} I(t) . \end{aligned} \quad (3.176)$$

This is a differential equation for $I(t)$ which has the solution (3.174) \square

As a consequence, the mean electric field amplitude of a coherent state evolves as

$$E(t) = \alpha \exp(|\alpha|^2(e^{i\kappa t} - 1)) \quad (3.177)$$

This result has some intriguing consequences. Let us first consider the case of a usual nonlinear crystal in which n_2 , and therefore κ , is small. In this case we can assume that $\kappa t \ll 1$ so that

$$E(t) \approx \alpha e^{i\kappa t |\alpha|^2} . \quad (3.178)$$

This is just the usual result for SPM. Note that the smallness of κt does not imply that the total phase shift $\Delta\phi_{\text{SPM}} = \text{Im}(|\alpha|^2(e^{i\kappa t} - 1)) \approx \kappa t |\alpha|^2$ needs to be small; for sufficiently large intensity $|\alpha|^2$ of the coherent signal field we can easily achieve phase shifts in the order of 1 rad or more.

On the other hand, if we consider a giant nonlinearity for a weak signal field the situation becomes very different. In this case we have $\Delta\phi_{\text{SPM}} = |\alpha|^2 \sin(\kappa t)$ which does not grow linearly in time. For a small value of $|\alpha|$ the SPM effect is small regardless of the size of κt ; neither a giant nonlinearity nor increasing the length of the nonlinear medium will result in an increased SPM phase shift. Even worse, the phase shift is unavoidably connected with a loss of fidelity because

$$|\exp(|\alpha|^2(e^{i\kappa t} - 1))| = \exp(|\alpha|^2(\cos(\kappa t) - 1)) < 1 . \quad (3.179)$$

This equation essentially states that in the presence of a large nonlinearity a coherent state will evolve into a very different state rather than just picking up an SPM phase shift.

This is seemingly in disagreement with experiments and has led to a number of theoretical investigations (until about the mid nineties) who are trying to adress this problem using a multi-mode description, i.e., a description that includes the dynamics of the quantum field operator. However, there seems to be no general agreement yet on the best way to describe Kerr media in QED.

As one example for a model, I will briefly discuss the approach taken by Boivin, Kärtner, and Haus [51]. The central ingredient of their theoretical model is to include a time-dependent nonlinear response rather than one which is constant in time. The corresponding Heisenberg equation of motion for $\mathcal{E}(z, t)$ takes the form

$$\partial_z \mathcal{E} = i\kappa n_{\text{NL}}(z, t) \mathcal{E}(z, t) \quad (3.180)$$

with

$$\kappa n_{\text{NL}}(z, t) = \int_{-\infty}^{\infty} dt' f(t') \mathcal{E}^\dagger(z, t - t') \mathcal{E}(z, t - t') + \hat{m}(z, t) , \quad (3.181)$$

where $f(t')$ describes a delayed Kerr-type nonlinearity and the operator \hat{m} incorporates the noise associated with the nonlinear medium.¹¹ It can be shown that this model predicts the usual SPM phase shift for a coherent state, regardless of the strength of the interaction. However, the associated noise inevitably leads to a loss of coherence which would make Kerr nonlinearities unsuitable to construct a controlled phase gate for quantum information [52]. So far this theoretical prediction has not been experimentally verified. Furthermore, the lack of agreement about which model properly describes Kerr media would call for applying alternative models to study this problem.

¹¹The reason why it is necessary to include a noise operator can be motivated by considering a damped harmonic oscillator with Heisenberg equation of motion $\partial_t a(t) = -\gamma a(t)$. The solution $a(t) = \exp(-\gamma t)a(0)$ would not fulfill the equal-time commutation relations $[a(t), a^\dagger(t)] = 1$ anymore. To cope with this one introduces a noise operator $m(t)$ which commutes with a, a^\dagger and fulfills $[m(t), m^\dagger(t')] = 2\gamma$. The modified dynamical equation, $\partial_t a(t) = -\gamma a(t) + m$, has the solution $a(t) = \exp(-\gamma t)a(0) + \int_0^t dt' \exp(-\gamma(t - t'))m(t')$. It is then easy to check that $[a(t), a^\dagger(t)] = 1$. While this may look like a mathematical trick, the noise operators have a concrete physical meaning: in any lossy medium the losses are generated by the interaction between the atoms of the medium and the radiation field. The noise operators are an effective description of the noise in the radiation field induced by the atoms.

Chapter 4

Quantum Mechanics of Atoms: a short Review

4.1 The Hydrogen Atom, Parity

Hamilton operator for a proton and an electron (V= Coulomb potential):

$$H = \frac{\mathbf{p}_p^2}{2M_p} + \frac{\mathbf{p}_e^2}{2m_e} + V(|\mathbf{x}_p - \mathbf{x}_e|) \quad (4.1)$$

$\mathbf{R} = \frac{1}{M_p + m_e}(M_p \mathbf{x}_p + m_e \mathbf{x}_e)$ is the center-of-mass coordinate, $\mathbf{r} = \mathbf{x}_e - \mathbf{x}_p$ the relative coordinate between the two particles, $M = M_p + m_e$, $\frac{1}{\mu} = \frac{1}{M_p} + \frac{1}{m_e}$. The Hamiltonian then can be rewritten as

$$H = \frac{\mathbf{P}^2}{2M} + \frac{\mathbf{p}^2}{2\mu} + V(|\mathbf{r}|) \quad (4.2)$$

In traditional atomic and molecular physics the relative motion is the central topic, in atom optics the center-of-mass motion.

Introduce *spherical coordinates* for the relative motion:

$$\Rightarrow \frac{\mathbf{p}^2}{2\mu} = -\frac{\hbar^2}{2\mu} \cdot \left(\frac{1}{r} \partial_r r \partial_r - \frac{\hat{\mathbf{L}}^2}{r^2} \right) \quad (4.3)$$

with $\mathbf{p} = -i\hbar\nabla$ and $\hat{\mathbf{L}}$ being the orbital angular momentum. One finds $[\hat{\mathbf{L}}, |\mathbf{r}|] = 0$. The eigenstates of H are therefore also eigenstates of L :

$$\begin{aligned} \psi &= Y_{lm}(\theta, \phi) f_l(r) \\ \hat{L}^2 Y_{lm} &= \hbar^2 l(l+1) Y_{lm} \end{aligned} \quad (4.4)$$

The radial part $f_l(r)$ of the wavefunction fullfills

$$Ef_l = \left(-\frac{\hbar^2}{2\mu} \frac{1}{r} \partial_r r \partial_r + \frac{\hbar^2}{2\mu} \frac{l(l+1)}{r^2} + V(r) \right) f_l \quad (4.5)$$

Solution: $f_{nl} \sim r^l \exp\left(-\frac{r}{na_0}\right) \cdot \text{Laguerre polynomials}$, $a_0 = \text{Bohr radius} \approx 0,5 \text{ \AA}$.

$$\Rightarrow \Psi_{nlm} = f_{nl} Y_{lm}(\theta, \phi) \iff |nlm\rangle \text{ (electronic state)} \quad (4.6)$$

Here n denotes the principal quantum number, l the orbital abgular momentum, and m the magnetic quantum number. There is one additional quantum number: *Spin* s of the electron, $s = \pm \frac{1}{2}$, $|\psi\rangle = |nlms\rangle$.

Parity:

$$\begin{aligned} \mathbf{r} &\longrightarrow -\mathbf{r} \\ |\mathbf{r}| &\longrightarrow |\mathbf{r}| \\ Y_{lm}(\theta, \phi) &\longrightarrow Y_{lm}(\pi - \theta, \phi + \pi) = (-1)^l Y_{lm}(\theta, \phi) \end{aligned} \quad (4.7)$$

\Rightarrow The parity of $|nlms\rangle$ is $(-1)^l$. Parity is very important for the dipole selection rules of atomic radiation.

4.2 Fine structure and spin

Of high practical relevance is the interaction of an atom with a static magnetic field,

$$H = -\boldsymbol{\mu} \cdot \mathbf{B} \quad (4.8)$$

where $\boldsymbol{\mu}$ is the magnetic dipole moment of the atom. A classical particle with angular momentum \mathbf{L} (e.g., on a circular orbit) and charge q possesses the magnetic moment

$$\boldsymbol{\mu} = \frac{q}{2M} \mathbf{L}. \quad (4.9)$$

The orbital contribution of the electron is

$$\boldsymbol{\mu}_e = -\frac{e}{2m_e} \hat{\mathbf{L}}. \quad (4.10)$$

For the proton we have because of $M_p \gg m_e$

$$\boldsymbol{\mu}_p = +\frac{e}{2M_p} \hat{\mathbf{L}} \ll \boldsymbol{\mu}_e. \quad (4.11)$$

Spin of the electron: ¹

$$-\frac{e}{2m_e}2\mathbf{S} = -\frac{\hbar e}{2m_e}\boldsymbol{\sigma}. \quad (4.12)$$

Fine structure is a consequence of the magnetic field which is created in the rest frame of the electron because of its motion around the nucleus. In the electrons rest frame the nucleus represents a moving charged particle, corresponding to an electric current that induces a magnetic field.

Rest frame of the nucleus: electric field $\mathbf{E} = -\nabla V_{Coul}$, $\mathbf{B} = 0$ (the nucleus creates no magnetic field as it is not moving in this frame).

Lorentz transformation to the electron's rest frame ²:

$$\begin{aligned} \mathbf{B}' &= \mathbf{v} \times \mathbf{E} \\ &= -\mathbf{v} \times \nabla V_{Coul} \\ &= -\frac{1}{M}\mathbf{p} \times \nabla V_{Coul} \end{aligned} \quad (4.13)$$

where \mathbf{v} is the velocity.

$$\begin{aligned} V_{Coul} &= V_{Coul}(|\mathbf{r}|) \implies \nabla V_{Coul} = \mathbf{e}_r \partial_r V_{Coul} = \mathbf{r} \frac{1}{r} \partial_r V_{Coul} \\ \implies \mathbf{B}' &= -\frac{1}{M} \left(\frac{1}{r} \partial_r V_{Coul} \right) \mathbf{p} \times \mathbf{r} = \frac{1}{M} \left(\frac{1}{r} \partial_r V_{Coul} \right) \mathbf{L}^{(e^-)} \end{aligned} \quad (4.14)$$

Thus, there exists a coupling between spin and orbital angular momentum of the electron:

$$\begin{aligned} H_{SB} &= -\mu_e \boldsymbol{\sigma} \mathbf{B}' \\ &= \mu_e \boldsymbol{\sigma} \cdot \mathbf{L} \left(\frac{1}{r} \partial_r V_{Coul} \right) \end{aligned} \quad (4.15)$$

An exact calculation of the matrix elements $\langle n'l'm's' | H_{SB} | nlm s \rangle$, including relativistic corrections, leads to

$$\begin{aligned} \Delta E_{FS} &= -\frac{1}{2} m_e c^2 (\alpha Z)^4 \frac{1}{h^3} \left(\frac{1}{j + \frac{1}{2}} - \frac{3}{4n} \right) \\ \Delta E_{FS} / \hbar &\approx 100 GHz \end{aligned} \quad (4.16)$$

¹The factor of 2, which appears here in contrast to the orbital angular momentum, is called gyromagnetic factor g . It follows from Dirac's equation for a relativistic electron that $g = 2$. However, vacuum fluctuations of the electromagnetic field cause small deviations from this value ($g \approx 2,002$).

²Actually the electron is moving on a non-inertial (circular) orbit and it is necessary to proceed differently. However, the result presented here agrees with the correct one up to a factor of 1/2 (Thomas factor) and explains the relevant physical ideas.

where $j = l \pm \frac{1}{2}$ denotes the total angular momentum of the electron.

Without relativistic corrections ΔE_{FS} would depend on l and s instead on $j = l + s$.

4.3 Hyperfine structure

Hyperfine structure is caused by two different effects:

1. Interaction between the electron's angular momentum and the nuclear spin.
2. QED corrections (vacuum fluctuations)

ad 1.: A magnetic dipole moment creates a magnetic field which other magnetic momenta can interact with. For the hydrogen atom the total angular momentum $\mathbf{j} = \mathbf{s} + \mathbf{l}$ of the electron creates a magnetic field at the position of the nucleus with which the latter's spin interacts.

Hyperfine spin:

$$\mathbf{F} = \mathbf{S}_k + \mathbf{j} \quad (4.17)$$

It has to be remarked that the addition of \mathbf{S}_k and \mathbf{j} to \mathbf{F} is *not fully equivalent* to the addition of the total spin $\mathbf{S}_G = \mathbf{S}_k + \mathbf{S}_e$ and the electronic orbital angular momentum \mathbf{L} to \mathbf{F} . This happens although the operators themselves are exactly equal. The reason is that with each of the two additions one associates a certain set of basis vectors diagonalizing \mathbf{F} . The two bases associated with $\mathbf{F} = \mathbf{S}_k + \mathbf{j}$ and $\mathbf{F} = \mathbf{S}_G + \mathbf{L}$ are different, see Appendix 10.2.

Chapter 5

The Interaction between Atoms and Light

5.1 Minimal Coupling

The classical Maxwell equations are gauge invariant,

$$\mathbf{A} \rightarrow \mathbf{A}' = \mathbf{A} + \nabla\chi \quad , \quad \phi \rightarrow \phi' = \phi - \partial_t\chi . \quad (5.1)$$

This invariance holds also in the presence of charge densities ϱ and currents \mathbf{j} and should also be present in the quantum description of electromagnetic fields.

5.1.1 Derivation of Minimal Coupling

The equation of motion for charged particles,

$$m\ddot{\mathbf{x}} = q\mathbf{E} + q\dot{\mathbf{x}} \times \mathbf{B} \quad \text{electric + Lorentz force}$$

can be derived from the Lagrangean ,

$$L = \frac{1}{2}m\dot{\mathbf{x}}^2 + q\dot{\mathbf{x}}\mathbf{A}(\mathbf{x}, t) - q\phi(\mathbf{x}, t) , \quad (5.2)$$

in the following way. The *canonically conjugated momentum* \mathbf{p} is defined by

$$p_i := \frac{\partial L}{\partial \dot{x}_i} \Rightarrow p_i = m\dot{x}_i + qA_i(\mathbf{x}, t) \quad (5.3)$$

Therefore, in the presence of an electromagnetic field the canonically conjugated momentum is different from the *kinetic momentum* $\mathbf{\Pi} = m\dot{\mathbf{x}}$. The Euler-Lagrange equations are given by

$$\frac{d}{dt}p_i = \frac{\partial L}{\partial x_i} \quad (5.4)$$

Insert (5.2) and (5.3):

$$\begin{aligned}\frac{\partial L}{\partial x_i} &= -q\partial_i\phi + q\dot{x}_k\partial_iA_k \\ \frac{d}{dt}p_i &= m\ddot{x}_i + q\frac{\partial}{\partial t}A_i(\mathbf{x}(t), t) + q\frac{\partial}{\partial x_k}A_i(\mathbf{x}(t), t)\dot{x}_k \\ \implies m\ddot{x}_i &= -q\partial_i\phi - q\partial_tA_i + q\dot{x}_k(\partial_iA_k - \partial_kA_i)\end{aligned}\quad (5.5)$$

verwenden $-\partial_i\phi - \partial_tA_i = E_i$ (by definition)

$$\implies m\ddot{x}_i = qE_i + q\dot{x}_k(\partial_iA_k - \partial_kA_i) \quad (5.6)$$

However, $B_j = (\text{curl } A)_j = \epsilon_{jmn}\partial_mA_n$

$$\begin{aligned}\implies \epsilon_{ikj}B_j &= \epsilon_{ikj}\epsilon_{jmn}\partial_mA_n \\ &= \epsilon_{jik}\epsilon_{jmn}\partial_mA_n \\ &= (\delta_{im}\delta_{kn} - \delta_{in}\delta_{km})\partial_mA_n \\ &= \partial_iA_k - \partial_kA_i \\ \implies m\ddot{x}_i &= qE_i + q\dot{x}_k\epsilon_{ikj}B_j \\ &= qE_i + q(\dot{\mathbf{x}} \times \mathbf{B})_i\end{aligned}\quad (5.7)$$

qed.

Using the Lagrange function one can derive the Hamiltonian and Schrödinger's equation: The classical Hamiltonian is defined by

$$H = p_i\dot{x}_i - L$$

Using $\dot{\mathbf{x}} = \frac{1}{m}(\mathbf{p} - q\mathbf{A})$ we want to write it as a function of \mathbf{p} and $\mathbf{x} \implies$

$$\begin{aligned}H &= p_i\frac{1}{m}(p_i - qA_i) - \frac{1}{2}m\frac{1}{m^2}(\mathbf{p} - q\mathbf{A})^2 - q\frac{1}{m}(\mathbf{p} - q\mathbf{A})\mathbf{A} + q\phi \\ H &= \frac{1}{2m}(\mathbf{p} - q\mathbf{A})^2 + q\phi\end{aligned}$$

The corresponding quantum mechanical Hamiltonian can be found by canonical quantisation:

$$\mathbf{x} \rightarrow \hat{\mathbf{x}} \quad ; \quad \mathbf{p} \rightarrow \hat{\mathbf{p}} \quad ; \quad [\hat{x}_i, \hat{p}_j] = \delta_{ij}$$

In position representation:

$$\mathbf{x} \rightarrow \hat{\mathbf{x}} \quad ; \quad \mathbf{p} \rightarrow -i\hbar\nabla$$

and therefore

$$\hat{H} = \frac{1}{2m}(-i\hbar\nabla - q\mathbf{A}(\hat{\mathbf{x}}, t))^2 + q\phi(\hat{\mathbf{x}}, t) \quad , \quad i\hbar\partial_t\psi(\mathbf{x}, t) = \hat{H}\psi(\mathbf{x}, t)$$

(5.8)

This is the Schrödinger equation for a non-relativistic particle in an electromagnetic field. The form $(\mathbf{p} - q\mathbf{A})$ is called *minimal coupling*.

5.1.2 Gauge Invariance of Minimal Coupling

Applying the transformation (5.1) to the Hamiltonian (5.8) one could be led to the conclusion that the Schrödinger equation is not gauge invariant:

$$\begin{aligned}\hat{H}' &= \frac{1}{2m} (-i\hbar\nabla - q\mathbf{A}(\hat{\mathbf{x}}, t) - q\nabla\chi)^2 + q\phi(\hat{\mathbf{x}}, t) - q\frac{\partial\chi}{\partial t}\psi \\ i\hbar\partial_t\psi(\mathbf{x}, t) &\neq \hat{H}\psi(\mathbf{x}, t)\end{aligned}$$

To restore the invariance under gauge transformations one also has to unitarily transform the wavefunction:

$$\boxed{\psi' = \psi \exp(iq\chi/\hbar)} \quad (5.9)$$

This implies

$$\begin{aligned}i\hbar\partial_t\psi' &= e^{iq\chi/\hbar} \left\{ i\hbar\frac{\partial\psi}{\partial t} - q\frac{\partial\chi}{\partial t}\psi \right\} \\ &= e^{iq\chi/\hbar} \left\{ \hat{H}\psi - q\frac{\partial\chi}{\partial t}\psi \right\} \\ &= e^{iq\chi/\hbar} \left\{ \frac{1}{2m} (-i\hbar\nabla - q\mathbf{A})^2 + q\phi \right\} \psi - q\frac{\partial\chi}{\partial t}\psi' \\ &= \left\{ \frac{1}{2m} (-i\hbar\nabla - q\mathbf{A} - q\nabla\chi)^2 + q\phi \right\} (e^{iq\chi/\hbar}\psi) - q\frac{\partial\chi}{\partial t}\psi' \\ &= \hat{H}'\psi'\end{aligned} \quad (5.10)$$

The minimal coupling scheme is of outstanding importance for high energy physics. Apart from gravity, all fundamental forces (that is, electromagnetic, weak, and strong interaction) do have the same basic structure. The simplest form of a gauge invariant theory is realized for the electromagnetic field. It is also called U(1) gauge theory because the transformation of the state (5.9) corresponds to a multiplication with a Unitary 1×1 matrix (= complex number of modulus 1). The other fundamental forces couple more fields. For example, the Special unitary SU(2) gauge theory of (electro-)weak interaction transforms the fields according to

$$\begin{pmatrix} \psi'_e \\ \psi'_{\nu_e} \end{pmatrix} = \exp(i\boldsymbol{\sigma} \cdot \boldsymbol{\chi}) \begin{pmatrix} \psi_e \\ \psi_{\nu_e} \end{pmatrix} \quad (5.11)$$

Here ψ_e is the wavefunction of the electron, ψ_{ν_e} that of the electron-neutrino, and $\boldsymbol{\sigma}$ are the Pauli matrices. The word *special* implies that the matrix $\exp(i\boldsymbol{\sigma} \cdot \boldsymbol{\chi})$ has determinant 1. The gauge potential for this theory can be written as $\mathbf{A} = \mathbf{A}^{(i)}\sigma_i$ and therefore consists of Three vector fields $\mathbf{A}^{(i)}$. These three fields do roughly

correspond to the W_{\pm} and the Z Boson which mediate the weak interaction between different particles, in the same way as photons do for the electromagnetic force.¹

5.2 The Power-Zienau-Woolley Transformation

Although minimal coupling is of highest importance for the description of the fundamental forces, it has several disadvantages when one wants to describe the interaction between atoms and light:

- For electrically neutral atoms which are composed of electrons, neutrons, and protons, minimal coupling is somewhat inconvenient. It would be better to describe the interaction using the atomic multipole moments.
- In minimal coupling the gauge potentials instead of the electric and magnetic field describe the coupling. Although this is not a principal problem a direct coupling to the physical fields would be manifestly gauge invariant and more intuitive.
- In Coulomb gauge the scalar potential ϕ can be identified with the unretarded Coulomb potential, since the latter solves Poisson's equation (which the equation for ϕ). The fact that the Coulomb potential is unretarded makes it appear as if minimal coupling would produce an instantaneous long-range force between the atoms.

The Power-Zienau-Woolley transformation (Refs. [28] and [29], see also [30] and [31]) is a unitary transformation of the minimal coupling. It leads to an equivalent description of the interaction, but with the above problems removed.

5.2.1 The Basic Idea

To derive the so-called dipole coupling² one can start from the fact that the Euler-Lagrange equations are not changed when one adds a total time derivative to the Lagrangian,

$$L'(x, \dot{x}, t) = L(x, \dot{x}, t) + \frac{d}{dt}F(x, t) \implies \frac{d}{dt} \frac{\partial L'}{\partial \dot{x}} - \frac{\partial L'}{\partial x} = \frac{d}{dt} \frac{\partial L}{\partial \dot{x}} - \frac{\partial L}{\partial x}$$

¹Actually the weak and the electromagnetic forces are entangled. Both together are described by a $U(1) \times SU(2)$ gauge theory. $U(1)$ thereby refers to hypercharge and not to the electromagnetic charge.

²A more precise name which is also used in the literature is *multipolar coupling* since the full Power-Zienau-Woolley transformation leads to a coupling of the electromagnetic field to all multipole moments of the atoms. However, in the applications one very often only takes the dipole moment into account. This is also the case for the simple example in this subsection.

For simplicity we consider here only constant fields \mathbf{E} and \mathbf{B} (for the general proof see below).

$$\begin{aligned}\mathbf{E} = \mathbf{E}_0; \mathbf{B} = \mathbf{B}_0 &\Longleftrightarrow \phi = -\mathbf{x}\mathbf{E}_0 \\ \mathbf{A} &= -\frac{1}{2}(\mathbf{x} \times \mathbf{B}_0)\end{aligned}$$

Choosing $F = -q\mathbf{x}\mathbf{A}$ one arrives at

$$\frac{d}{dt}F = -q\dot{\mathbf{x}}\mathbf{A} + \frac{q}{2}(\mathbf{x} \times \dot{\mathbf{x}})\mathbf{B}_0 \quad (5.12)$$

$$L' = \underbrace{\frac{1}{2}m\dot{\mathbf{x}}^2 + q\dot{\mathbf{x}}\mathbf{A} + q\mathbf{x}\mathbf{E}_0 - q\dot{\mathbf{x}}\mathbf{A}}_L + \frac{q}{2}(\mathbf{x} \times \dot{\mathbf{x}})\mathbf{B}_0 \quad (5.13)$$

$$= \frac{1}{2}m\dot{\mathbf{x}}^2 + q\mathbf{x}\mathbf{E}_0 + \frac{q}{2m}\mathbf{L}\mathbf{B}_0 \quad \text{mit } \mathbf{L} = (\mathbf{x} \times m\dot{\mathbf{x}}) \quad (5.14)$$

Setting $\mathbf{d} := q\mathbf{x}$ electric dipole moment of the particle one finds for the new
 $\boldsymbol{\mu} := \frac{q}{2m}\mathbf{L}$ magnetic moment of the particle
 Lagrangean

$$L' = \frac{1}{2}m\dot{\mathbf{x}}^2 + \mathbf{d}\mathbf{E}_0 + \boldsymbol{\mu}\mathbf{B}_0$$

Advantage: Now the canonical momentum agrees with the kinetic momentum, $\mathbf{p} = m\dot{\mathbf{x}}$. In addition, the particle now couples to the physical fields and therefore is manifestly gauge invariant. It also involves the dipole momenta which are better suited for electrically neutral particles.

5.2.2 The Complete Transformation

We consider an ensemble of point particles with charges q_α and positions \mathbf{x}_α . Their charge density is given by

$$\varrho(\mathbf{x}) = \sum_{\alpha} q_{\alpha} \delta(\mathbf{x} - \mathbf{x}_{\alpha}) \quad (5.15)$$

Since we are particularly interested in the behaviour of a gas of neutral atoms, we assume that the total charge is zero, $\sum_{\alpha} q_{\alpha} = 0$. The polarization of macroscopic Electrodynamics fullfills $\text{div} \mathbf{P} = -\langle \varrho_{\text{geb}} \rangle$ (see Sec. ??). For the present case its solution is given by

$$\mathbf{P}(\mathbf{x}) = \sum_{\alpha} \int_0^1 du \, q_{\alpha} (\mathbf{x}_{\alpha} - \mathbf{R}) \delta(\mathbf{x} - \mathbf{R} - u(\mathbf{x}_{\alpha} - \mathbf{R})) \quad (5.16)$$

Proof:

$$\begin{aligned}
 \partial_i P_i(\mathbf{x}) &= \sum_{\alpha} \int_0^1 du \, q_{\alpha}(\mathbf{x}_{\alpha} - \mathbf{R})_i \partial_i \delta(\mathbf{x} - \mathbf{R} - u(\mathbf{x}_{\alpha} - \mathbf{R})) \\
 &= \sum_{\alpha} \int_0^1 du \, q_{\alpha}(\mathbf{x}_{\alpha} - \mathbf{R})_i \delta'(\mathbf{x}_i - \mathbf{R}_i - u(\mathbf{x}_{\alpha} - \mathbf{R})_i) \times \\
 &\quad \delta(\mathbf{x}_j - \mathbf{R}_j - u(\mathbf{x}_{\alpha} - \mathbf{R})_j) \delta(\mathbf{x}_k - \mathbf{R}_k - u(\mathbf{x}_{\alpha} - \mathbf{R})_k)
 \end{aligned}$$

where we sum over i and $(i, j, k) = (1, 2, 3), (2, 3, 1), (3, 1, 2)$. δ' is the derivative of the δ -distribution. We have

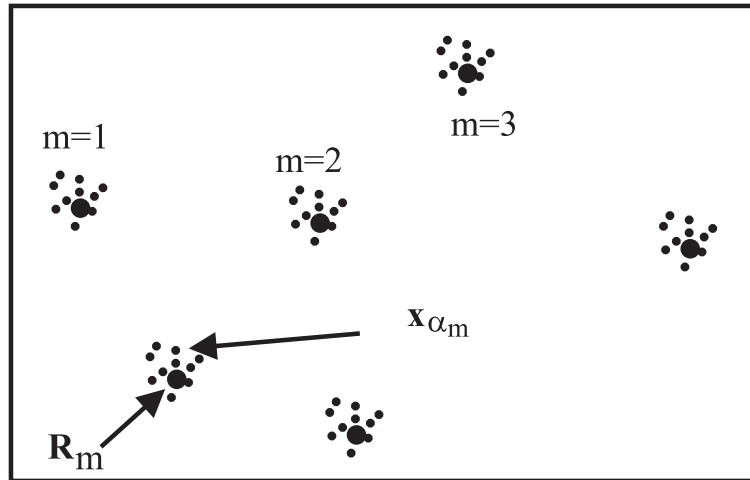
$$(\mathbf{x}_{\alpha} - \mathbf{R})_i \delta'(\mathbf{x}_i - \mathbf{R}_i - u(\mathbf{x}_{\alpha} - \mathbf{R})_i) = -\partial_u \delta(\mathbf{x}_i - \mathbf{R}_i - u(\mathbf{x}_{\alpha} - \mathbf{R})_i) \quad (5.18)$$

and hence

$$\begin{aligned}
 \partial_i P_i(\mathbf{x}) &= -\sum_{\alpha} \int_0^1 du \, q_{\alpha} \partial_u \delta(\mathbf{x} - \mathbf{R} - u(\mathbf{x}_{\alpha} - \mathbf{R})) \\
 &= -\sum_{\alpha} q_{\alpha} \{\delta(\mathbf{x} - \mathbf{x}_{\alpha}) - \delta(\mathbf{x} - \mathbf{R})\} \\
 &= Q \delta(\mathbf{x} - \mathbf{R}) - \sum_{\alpha} q_{\alpha} \delta(\mathbf{x} - \mathbf{x}_{\alpha}) \quad (5.19)
 \end{aligned}$$

The total charge Q vanishes for neutral atoms, and the second term corresponds to the charge density q.e.d.

To understand all consequences of the Power-Zienau-Woolley transformation, one needs to consider a gas with more than a single atom or molecule. We therefore consider a charge distribution in which the charge carriers are grouped into electrically neutral atoms or molecules. The nucleus of the m th atom is placed at a position \mathbf{R}_m , and the particle coordinates (electrons + nucleus) are denoted by \mathbf{x}_{α_m} .



In minimal coupling, the complete Hamiltonian of this system is given by

$$H = \sum_m \sum_{\alpha_m} \frac{1}{2M_{\alpha_m}} (\mathbf{p}_{\alpha_m} - q_{\alpha_m} \mathbf{A}(\mathbf{x}_{\alpha_m}))^2 + \frac{\varepsilon_0}{2} \int d^3x ((\mathbf{E}^\perp)^2 + c^2 \mathbf{B}^2) + \sum_{m < m'} V_{\text{Coul}}(m, m') + \sum_m V_{\text{Coul}}(m). \quad (5.20)$$

Here, \mathbf{E}^\perp is the operator of the transverse electric field. It is the same operator which we up to now (in absence of charges) simply have denoted by \mathbf{E} . In the presence of charges, the complete electric field takes the form $\mathbf{E} = \mathbf{E}^\perp - \nabla V_{\text{Coul}}(\mathbf{x})$. The Coulomb interaction between the particles of one atom is given by

$$V_{\text{Coul}}(m) = \sum_{\alpha_m < \beta_m} \frac{q_{\alpha_m} q_{\beta_m}}{4\pi\varepsilon_0 |\mathbf{x}_{\alpha_m} - \mathbf{x}_{\beta_m}|}. \quad (5.21)$$

On the other hand, the interaction between the particles of two different atoms can be written as

$$V_{\text{Coul}}(m, m') = \sum_{\alpha_m, \beta_{m'}} \frac{q_{\alpha_m} q_{\beta_{m'}}}{4\pi\varepsilon_0 |\mathbf{x}_{\alpha_m} - \mathbf{x}_{\beta_{m'}}|}. \quad (5.22)$$

To construct the unitary transformation that connects minimal coupling to multipolar coupling, we employ the total polarization of the gas, which is a sum over the polarization of the individual atoms:

$$\mathbf{P}(\mathbf{x}) = \sum_m \mathbf{P}_m(\mathbf{x}) \quad (5.23)$$

$$\mathbf{P}_m(\mathbf{x}) = \sum_{\alpha_m} \int_0^1 du q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \delta(\mathbf{x} - \mathbf{R}_m - u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \quad (5.24)$$

The *Power-Zienau-Woolley-Transformation* is then given by

$$U = \exp \left(\frac{i}{\hbar} \int d^3x \mathbf{P}(\mathbf{x}) \cdot \mathbf{A}(\mathbf{x}) \right) \quad (5.25)$$

Explicit expressions for the transformation

Our task is to find explicit expressions for the transformed Hamiltonian and other operators of relevance. Since U is a function of \mathbf{x}_{α_m} and $\mathbf{A}(\mathbf{x})$, it is obvious that because $[\mathbf{A}_i(\mathbf{x}), \mathbf{A}_j(\mathbf{x}')] = [\mathbf{A}_i(\mathbf{x}), \mathbf{B}_j(\mathbf{x}')] = 0$ the vector potential, the magnetic field and the particle coordinates as well as the polarization transform

trivially,

$$\tilde{\mathbf{A}} = U^\dagger \mathbf{A} U = \mathbf{A} \quad (5.26)$$

$$\tilde{\mathbf{B}} = \mathbf{B} \quad (5.27)$$

$$\tilde{\mathbf{x}}_{\alpha_m} = \mathbf{x}_{\alpha_m} \quad (5.28)$$

$$\tilde{\mathbf{P}} = \mathbf{P} . \quad (5.29)$$

All other fields can be treated by using the theorem $\exp(-iS)E\exp(iS) = E - i[S, E] + \dots$. We find for the electric field

$$\tilde{\mathbf{E}}_i^\perp(\mathbf{x}) = \mathbf{E}_i^\perp(\mathbf{x}) - \frac{i}{\hbar} \int d^3x' \mathbf{P}_j(\mathbf{x}') [\mathbf{A}_j(\mathbf{x}'), \mathbf{E}_i^\perp(\mathbf{x})] \quad (5.30)$$

$$= \mathbf{E}_i^\perp(\mathbf{x}) - \frac{i}{\hbar} \int d^3x' \mathbf{P}_j(\mathbf{x}') \frac{-i\hbar}{\varepsilon_0} \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') . \quad (5.31)$$

Recalling that the transverse (source-free) part of a vector field \mathbf{P} is given by $(T\mathbf{P})_i(\mathbf{x}) = \int d^3x' \mathbf{P}_j(\mathbf{x}') \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}')$, we can derive

$$\tilde{\mathbf{E}}^\perp(\mathbf{x}) = \mathbf{E}^\perp(\mathbf{x}) - \frac{1}{\varepsilon_0} \mathbf{P}^\perp(\mathbf{x}) . \quad (5.32)$$

This equation has some interesting consequences. If we write it in the form

$$\varepsilon_0 \mathbf{E}^\perp(\mathbf{x}) = \varepsilon_0 \tilde{\mathbf{E}}^\perp(\mathbf{x}) + \tilde{\mathbf{P}}^\perp(\mathbf{x}) = \tilde{\mathbf{D}}^\perp \quad (5.33)$$

it becomes obvious that the *old* operator $\varepsilon_0 \mathbf{E}^\perp$ of the electric field *in the new picture* plays the role of the dielectric displacement field $\tilde{\mathbf{D}}^\perp$. In other words: if one uses minimal coupling, the electric field is given by \mathbf{E}^\perp and the dielectric displacement by $\varepsilon_0 \mathbf{E}^\perp + \mathbf{P}^\perp$. In dipole coupling, the electric field is given by $\tilde{\mathbf{E}}^\perp$ and the dielectric displacement by $\varepsilon_0 \tilde{\mathbf{E}}^\perp + \tilde{\mathbf{P}}^\perp = \varepsilon_0 \mathbf{E}^\perp$. Since the operator \mathbf{E}^\perp has a simple form, it is also used in multipolar coupling to express the interaction.

it remains to calculate the transformation of the particle momenta. One easily finds

$$\begin{aligned} \tilde{\mathbf{p}}_{\alpha_m} &= \mathbf{p}_{\alpha_m} - i[S, \mathbf{p}_{\alpha_m}] + \dots \\ &= \mathbf{p}_{\alpha_m} + \hbar \nabla_{\alpha_m} S . \end{aligned} \quad (5.34)$$

The calculation of the gradient will be done below. In summary we obtain for the transformed Hamiltonian the expression

$$\begin{aligned} \tilde{H} &= \sum_m \sum_{\alpha_m} \frac{1}{2M_{\alpha_m}} (\mathbf{p}_{\alpha_m} + \hbar \nabla_{\alpha_m} S - q_{\alpha_m} \mathbf{A}(\mathbf{x}_{\alpha_m}))^2 \\ &\quad + \frac{\varepsilon_0}{2} \int d^3x \left(\frac{1}{\varepsilon_0^2} (\tilde{\mathbf{D}}^\perp - \mathbf{P}^\perp)^2 + c^2 \mathbf{B}^2 \right) + \\ &\quad \sum_{m < m'} V_{\text{Coul}}(m, m') + \sum_m V_{\text{Coul}}(m) \end{aligned} \quad (5.35)$$

Calculation of the polarization integrals

This expression can be considerably simplified. We start with the second line:

$$\begin{aligned} \frac{1}{2\varepsilon_0} \int d^3x (\tilde{\mathbf{D}}^\perp - \mathbf{P}^\perp)^2 &= \frac{1}{2\varepsilon_0} \int d^3x \left((\tilde{\mathbf{D}}^\perp)^2 - 2\tilde{\mathbf{D}}^\perp \cdot \mathbf{P}^\perp + (\mathbf{P}^\perp)^2 \right) \\ &= \frac{1}{2\varepsilon_0} \int d^3x \left((\tilde{\mathbf{D}}^\perp)^2 - 2\tilde{\mathbf{D}}^\perp \cdot \mathbf{P}^\perp + (\mathbf{P} - \mathbf{P}^\parallel)^2 \right), \end{aligned} \quad (5.36)$$

where \mathbf{P}^\parallel denotes the longitudinal part of the vector field (i.e., the Fourier transform of the field is parallel to the wave vector, not transverse). We can now make use of the following relation,

$$\int d^3x \mathbf{R}^\perp(\mathbf{x}) \cdot \mathbf{S}(\mathbf{x}) = \int d^3x \mathbf{R}^\perp(\mathbf{x}) \cdot \mathbf{S}^\perp(\mathbf{x}). \quad (5.37)$$

To prove it we first show that the transversalization operator is idempotent. By definition, we have

$$(T^2 R)_i(\mathbf{x}) = \int d^3x' \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \int d^3x'' \delta_{jk}^\perp(\mathbf{x}' - \mathbf{x}'') \mathbf{R}_k(\mathbf{x}''). \quad (5.38)$$

With

$$\begin{aligned} \int d^3x' \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \delta_{jk}^\perp(\mathbf{x}' - \mathbf{x}'') &= \int d^3x' \int \frac{d^3k}{(2\pi)^3} \frac{d^3k'}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}')} e^{i\mathbf{k}' \cdot (\mathbf{x}' - \mathbf{x}'')} \times \\ &\quad \left(\delta_{ij} - \frac{\mathbf{k}_i \mathbf{k}_j}{\mathbf{k}^2} \right) \left(\delta_{jk} - \frac{\mathbf{k}'_j \mathbf{k}'_k}{(\mathbf{k}')^2} \right) \\ &= \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}'')} \left(\delta_{ij} - \frac{\mathbf{k}_i \mathbf{k}_j}{\mathbf{k}^2} \right) \left(\delta_{jk} - \frac{\mathbf{k}_j \mathbf{k}_k}{\mathbf{k}^2} \right) \\ &= \delta_{ik}^\perp(\mathbf{x} - \mathbf{x}'') \end{aligned} \quad (5.39)$$

it follows that

$$\begin{aligned} (T^2 R)_i(\mathbf{x}) &= \int d^3x'' \delta_{ik}^\perp(\mathbf{x} - \mathbf{x}'') \mathbf{R}_k(\mathbf{x}'') \\ &= (TR)_i(\mathbf{x}) \end{aligned} \quad (5.40)$$

In addition, T is Hermitean with respect to the standard scalar product:

$$\begin{aligned} \int d^3x \mathbf{R}(\mathbf{x}) \cdot (T\mathbf{S})(\mathbf{x}) &= \int d^3x \mathbf{R}_i(\mathbf{x}) \int d^3x' \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}') \mathbf{S}_j(\mathbf{x}') \\ &= \int d^3x' (T\mathbf{R})(\mathbf{x}') \cdot \mathbf{S}(\mathbf{x}') \end{aligned} \quad (5.41)$$

Relation (5.37) then follows from

$$\begin{aligned}
 \int d^3x \mathbf{R}^\perp(\mathbf{x}) \cdot \mathbf{S}(\mathbf{x}) &= \int d^3x (T\mathbf{R})(\mathbf{x}) \cdot \mathbf{S}(\mathbf{x}) \\
 &= \int d^3x (T^2\mathbf{R})(\mathbf{x}) \cdot \mathbf{S}(\mathbf{x}) \\
 &= \int d^3x (T\mathbf{R})(\mathbf{x}) \cdot (T\mathbf{S})(\mathbf{x}) \\
 &= \int d^3x \mathbf{R}^\perp(\mathbf{x}) \cdot \mathbf{S}^\perp(\mathbf{x}) . \tag{5.42}
 \end{aligned}$$

Eq. (5.37) is helpful to simplify the integral over the polarization:

$$\begin{aligned}
 \frac{1}{2\varepsilon_0} \int d^3x \left(\mathbf{P}^2 - 2\mathbf{P} \cdot \mathbf{P}^\parallel + (\mathbf{P}^\parallel)^2 \right) &= \frac{1}{2\varepsilon_0} \int d^3x \left(\mathbf{P}^2 - 2(\mathbf{P}^\parallel)^2 + (\mathbf{P}^\parallel)^2 \right) \\
 &= \frac{1}{2\varepsilon_0} \int d^3x \left(\mathbf{P}^2 - (\mathbf{P}^\parallel)^2 \right) \tag{5.43}
 \end{aligned}$$

The integration over the longitudinal part can be calculated exactly. Since the source of this term is the charge distribution, $\text{div}\mathbf{P} = \text{div}\mathbf{P}^\parallel = -\varrho$, it is reasonable that it is related to the Coulomb potential. To demonstrate this we first rewrite the latter in the form

$$\begin{aligned}
 V_{\text{Coul}}(\mathbf{x}) &= \frac{1}{4\pi\varepsilon_0} \int d^3x' \frac{\varrho(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} \\
 &= \frac{-1}{4\pi\varepsilon_0} \int d^3x' \frac{\text{div}\mathbf{P}^\parallel(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} \\
 &= \frac{1}{4\pi\varepsilon_0} \int d^3x' \mathbf{P}_i^\parallel(\mathbf{x}') \partial'_i \frac{1}{|\mathbf{x} - \mathbf{x}'|} \tag{5.44}
 \end{aligned}$$

The gradient of the Coulomb potential, which simply is the negative longitudinal electric field, we find

$$\begin{aligned}
 \partial_j V_{\text{Coul}}(\mathbf{x}) &= \frac{1}{4\pi\varepsilon_0} \int d^3x' \mathbf{P}_i^\parallel(\mathbf{x}') \partial_j \partial'_i \frac{1}{|\mathbf{x} - \mathbf{x}'|} \\
 &= \frac{-1}{4\pi\varepsilon_0} \int d^3x' \mathbf{P}_i^\parallel(\mathbf{x}') \partial_j \partial'_i \frac{1}{|\mathbf{x} - \mathbf{x}'|} \tag{5.45}
 \end{aligned}$$

To calculate the double derivative of the integrand, we use the Fourier transform

$$\int d^3x \frac{e^{-i\mathbf{k} \cdot \mathbf{x}}}{|\mathbf{x}|} = \frac{4\pi}{\mathbf{k}^2} \tag{5.46}$$

To prove this one can use spherical coordinates, with the z-axis along \mathbf{k} . One then finds ³

$$\begin{aligned}
 \int d^3x \frac{e^{-i\mathbf{k}\cdot\mathbf{x}}}{|\mathbf{x}|} &= \int_0^\infty r^2 dr \int d\cos\vartheta \int d\varphi \frac{e^{-ikr\cos\vartheta}}{r} \\
 &= 2\pi \int_0^\infty r dr \frac{1}{-ikr} (e^{-ikr} - e^{ikr}) \\
 &= \frac{2\pi i}{k} \left\{ \pi\delta(k) - i\frac{\mathcal{P}}{k} - \left(\pi\delta(k) + i\frac{\mathcal{P}}{k} \right) \right\} \\
 &= \frac{4\pi}{k^2}
 \end{aligned} \tag{5.47}$$

We thus find for the double derivative

$$\begin{aligned}
 \partial_j \partial_i \frac{1}{|\mathbf{x} - \mathbf{x}'|} &= \partial_j \partial_i \int \frac{d^3k}{(2\pi)^3} \frac{4\pi e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}}{k^2} \\
 &= -4\pi \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')} \frac{\mathbf{k}_i \mathbf{k}_j}{k^2} \\
 &= -4\pi (\delta_{ij} \delta(\mathbf{x} - \mathbf{x}') - \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}'))
 \end{aligned} \tag{5.48}$$

so that

$$\begin{aligned}
 \partial_j V_{\text{Coul}}(\mathbf{x}) &= \frac{-1}{4\pi\epsilon_0} \int d^3x' \mathbf{P}_i^\parallel(\mathbf{x}') (-4\pi) (\delta_{ij} \delta(\mathbf{x} - \mathbf{x}') - \delta_{ij}^\perp(\mathbf{x} - \mathbf{x}')) \\
 &= \frac{1}{\epsilon_0} \mathbf{P}_j^\parallel(\mathbf{x})
 \end{aligned} \tag{5.49}$$

The longitudinal electric field is therefore completely characterized by the polarization, so that

$$\begin{aligned}
 -\frac{1}{2\epsilon_0} \int d^3x (\mathbf{P}^\parallel)^2 &= -\frac{\epsilon_0}{2} \int d^3x (\nabla V_{\text{Coul}})^2 \\
 &= \frac{\epsilon_0}{2} \int d^3x V_{\text{Coul}} \Delta V_{\text{Coul}} \\
 &= -\frac{1}{2} \int d^3x V_{\text{Coul}} \varrho,
 \end{aligned} \tag{5.50}$$

³The omission of the principal part \mathcal{P} in the last line is based on the following considerations. Like every distribution, \mathcal{P}/k is only defined as an integrand and needs to be multiplied with a test function (which has to be infinitely often differentiable, normalizable, without poles). In the second-to-last row, an additional factor of $1/k$ appears, which is not a test function. Therefore, the result is only properly defined if one considers test functions which go at least linearly against zero for $k \rightarrow 0$. In theoretical physics, however, it is customary to use this equation without caring about test functions. The principal value is then meaningless, since it does not result in a finite value if the divergence is of the form $1/k^2$. It is therefore simply omitted.

where the last line follows from the Coulomb potential obeying Poisson's equation. Inserting Eq. (5.15) for the charge density, one arrives at

$$-\frac{1}{2\varepsilon_0} \int d^3x (\mathbf{P}^{\parallel})^2 = -\frac{1}{2} \sum_{m, \alpha_m} \sum_{m', \alpha'_{m'}} \frac{q_{\alpha_m} q_{\alpha'_{m'}}}{4\pi\varepsilon_0 |\mathbf{x}_{\alpha_m} - \mathbf{x}_{\alpha'_{m'}}|} \quad (5.51)$$

In this expression, we have to remove the terms with indential particles since they diverge. The rest is symmetric under exchange of primed and unprimed indices, so that

$$\begin{aligned} -\frac{1}{2\varepsilon_0} \int d^3x (\mathbf{P}^{\parallel})^2 &= -\sum_{m < m'} \sum_{\alpha_m, \alpha'_{m'}} \frac{q_{\alpha_m} q_{\alpha'_{m'}}}{4\pi\varepsilon_0 |\mathbf{x}_{\alpha_m} - \mathbf{x}_{\alpha'_{m'}}|} \\ &\quad - \sum_m \sum_{\alpha_m < \alpha'_m} \frac{q_{\alpha_m} q_{\alpha'_m}}{4\pi\varepsilon_0 |\mathbf{x}_{\alpha_m} - \mathbf{x}_{\alpha'_m}|} \\ &= -\sum_{m < m'} V(m, m') - \sum_m V(m) \end{aligned} \quad (5.52)$$

The integral over the longitudinal part of the polarization therefore exactly cancels the Coulomb interaction between the atoms. This is the reason why we decomposed the polarization into the complete and the longitudinal polarization in Eq. (5.36). The remaining integral over the complete polarization field can now be decomposed into the individual contribution of each atom,

$$\int d^3x \mathbf{P}^2 = \sum_{m, m'} \int d^3x \mathbf{P}_m \mathbf{P}_{m'} \quad (5.53)$$

In Eq. (5.24) one can see that the polarization of the m th atom is strongly localized around the atom itself. Since in an atomic gas the distance between the atoms is much larger then their size, one can neglect terms with $m \neq m'$ and thus deduces

$$\begin{aligned} \frac{1}{2\varepsilon_0} \int d^3x \mathbf{P}^2 &\approx \frac{1}{2\varepsilon_0} \sum_m \int d^3x \mathbf{P}_m^2 \\ &= \frac{1}{2\varepsilon_0} \sum_m \int d^3x \left((\mathbf{P}_m^{\perp})^2 + (\mathbf{P}_m^{\parallel})^2 \right) \\ &= \frac{1}{2\varepsilon_0} \sum_m \int d^3x (\mathbf{P}_m^{\perp})^2 + \sum_m V(m) \end{aligned} \quad (5.54)$$

Hamiltonian and multipolar expansion

Inserting all calculations in the transformed Hamiltonian (5.35) one obtains

$$\tilde{H} = \sum_m H_m + H_{\text{rad}} + H_{\text{int}} \quad (5.55)$$

$$H_m = \sum_{\alpha_m} \frac{\mathbf{p}_{\alpha_m}^2}{2M_{\alpha_m}} + V(m) + \frac{1}{2\varepsilon_0} \int d^3x (\mathbf{P}_m^\perp)^2 \quad (5.56)$$

$$H_{\text{rad}} = \frac{1}{2\varepsilon_0} \int d^3x (\mathbf{D}^\perp)^2 + \frac{1}{2\mu_0} \int d^3x \mathbf{B}^2 \quad (5.57)$$

$$\begin{aligned} H_{\text{int}} = & -\frac{1}{\varepsilon_0} \int d^3x \mathbf{D}^\perp \cdot \mathbf{P} \\ & + \sum_{m, \alpha_m} \frac{1}{2M_{\alpha_m}} (\mathbf{n}_{\alpha_m} \cdot \mathbf{p}_{\alpha_m} + \mathbf{p}_{\alpha_m} \cdot \mathbf{n}_{\alpha_m} + (\mathbf{n}_{\alpha_m})^2) \end{aligned} \quad (5.58)$$

with

$$\begin{aligned} (\mathbf{n}_{\alpha_m})_i &:= \hbar(\nabla_{\alpha_m})_i S - q_{\alpha_m} \mathbf{A}_i(\mathbf{x}_{\alpha_m}) \quad (5.59) \\ &= \hbar(\nabla_{\alpha_m})_i \int_0^1 du q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \cdot \mathbf{A}(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) - q_{\alpha_m} \mathbf{A}_i(\mathbf{x}_{\alpha_m}) \\ &= \int_0^1 du q_{\alpha_m} \left(\mathbf{A}_i(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \right. \\ &\quad \left. + (\mathbf{x}_{\alpha_m} - \mathbf{R}_m)_j u \partial_i \mathbf{A}_j(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \right) - q_{\alpha_m} \mathbf{A}_i(\mathbf{x}_{\alpha_m}) \end{aligned}$$

The vector \mathbf{n}_{α_m} can be brought into a form which is explicitly independent from the potential. To do so one uses

$$\int_0^1 du u \partial_u \mathbf{A}(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) = \mathbf{A}(\mathbf{x}_{\alpha_m}) - \int_0^1 du \mathbf{A}(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \quad (5.60)$$

which can be proven by partial integration. Introducing the notation $\mathbf{r}_{\alpha_m} := \mathbf{x}_{\alpha_m} - \mathbf{R}_m$ it follows that

$$\begin{aligned} (\mathbf{n}_{\alpha_m})_i &:= \int_0^1 du q_{\alpha_m} (\mathbf{A}_i(\mathbf{R}_m + u\mathbf{r}_{\alpha_m}) + (\mathbf{r}_{\alpha_m})_j u \partial_i \mathbf{A}_j(\mathbf{R}_m + u\mathbf{r}_{\alpha_m})) \\ &\quad - \int_0^1 du q_{\alpha_m} (u \partial_u \mathbf{A}_i(\mathbf{R}_m + u\mathbf{r}_{\alpha_m}) + \mathbf{A}_i(\mathbf{R}_m + u\mathbf{r}_{\alpha_m})) \\ &= \int_0^1 du q_{\alpha_m} ((\mathbf{r}_{\alpha_m})_j u \partial_i \mathbf{A}_j(\mathbf{R}_m + u\mathbf{r}_{\alpha_m}) - u(\mathbf{r}_{\alpha_m})_j \partial_j \mathbf{A}_i(\mathbf{R}_m + u\mathbf{r}_{\alpha_m})) \\ &= \int_0^1 du q_{\alpha_m} u (\mathbf{r}_{\alpha_m})_j (\partial_i \mathbf{A}_j(\mathbf{R}_m + u\mathbf{r}_{\alpha_m}) - \partial_j \mathbf{A}_i(\mathbf{R}_m + u\mathbf{r}_{\alpha_m})) \end{aligned} \quad (5.61)$$

and therefore, because of $\partial_i \mathbf{A}_j - \partial_j \mathbf{A}_i = \varepsilon_{ijk} \mathbf{B}_k$,

$$\mathbf{n}_{\alpha_m} = q_{\alpha_m} \int_0^1 u \, du \, (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \times \mathbf{B}(\mathbf{R}_m + u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \quad (5.62)$$

This is the final form of the multipolar Hamiltonian. We emphasize again that no approximation has been made apart from the assumption that the total polarizations of two different molecules have little overlap. It is therefore fully justified to use this coupling instead of minimal coupling. The most important features of the Hamiltonian (5.55) are as follows.

- It is explicitly gauge invariant since it only contains the electric and the magnetic field.
- The Coulomb potential between different atoms has disappeared. The interaction between the atoms is now exclusively mediated through the retarded dynamical fields $\tilde{\mathbf{D}}^\perp$ and \mathbf{B} .

5.3 Dipole approximation and dipole coupling

The typical size r of an atom is in the order of Bohr's radius ($\approx 0.5 \times 10^{-10}$ m). Since optical wavelengths $\lambda = 2\pi/k$ are in the range of several hundreds of nm, it is justified to restrict the multipolar expansion of the interaction Hamiltonian after the first few terms. The reason is that multipole moments of order l are proportional to $(kr)^l$ and $kr \ll 1$ is valid, see Eq. (1.18). We therefore keep only the lowest order terms and obtain for the polarization

$$\begin{aligned} \mathbf{P}(\mathbf{x}) &= \sum_m \mathbf{P}_m(\mathbf{x}) \\ &= \sum_m \sum_{\alpha_m} q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \int_0^1 du \, \delta(\mathbf{x} - \mathbf{R}_m - u(\mathbf{x}_{\alpha_m} - \mathbf{R}_m)) \\ &\approx \sum_m \sum_{\alpha_m} q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \int_0^1 du \, \delta(\mathbf{x} - \mathbf{R}_m) + O(|\mathbf{x}_{\alpha_m} - \mathbf{R}_m|^2) \\ &= \sum_m \delta(\mathbf{x} - \mathbf{R}_m) \sum_{\alpha_m} q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \\ &= \sum_m \delta(\mathbf{x} - \mathbf{R}_m) \mathbf{d}_m, \end{aligned} \quad (5.63)$$

where \mathbf{d}_m is the dipole moment of the m th atom/molecule. Analogously one finds for the second coupling term

$$\mathbf{n}_{\alpha_m} \approx q_{\alpha_m} \int_0^1 u \, du \, (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \times \mathbf{B}(\mathbf{R}_m) + O(|\mathbf{x}_{\alpha_m} - \mathbf{R}_m|^2) \quad (5.64)$$

and thus

$$\begin{aligned} \mathbf{n}_{\alpha_m} \cdot \mathbf{p}_{\alpha_m} + \mathbf{p}_{\alpha_m} \cdot \mathbf{n}_{\alpha_m} + (\mathbf{n}_{\alpha_m})^2 &\approx 2q_{\alpha_m} \mathbf{p}_{\alpha_m} \cdot \int_0^1 u \, du (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \times \mathbf{B}(\mathbf{R}_m) \\ &= -q_{\alpha_m} \mathbf{L}_{\alpha_m} \cdot \mathbf{B}(\mathbf{R}_m) \end{aligned} \quad (5.65)$$

This is the *dipole approximation*. The associated interaction Hamiltonian is called *dipole coupling* and given by

$$\boxed{\tilde{H}_{\text{int}} = -\frac{1}{\varepsilon_0} \sum_m \mathbf{d}_m \cdot \tilde{\mathbf{D}}^\perp(\mathbf{R}_m) - \sum_m \boldsymbol{\mu}_m \cdot \mathbf{B}(\mathbf{R}_m)} \quad (5.66)$$

where $\boldsymbol{\mu}_m = \sum_{\alpha_m} q_{\alpha_m} \mathbf{L}_{\alpha_m} / (2M_{\alpha_m})$ is the magnetic dipole moment that is associated with the orbital angular momentum of the electrons. The spin contribution simply doesn't appear here because it is omitted in our treatment of the Power-Zienau-Woolley transformation. It would of course be present if we start from particles with spin.

We remark that only the complete multipolar Hamiltonian is unitarily equivalent to the minimal coupling scheme. As soon as approximations are made, the use of the different interaction forms leads to slightly different results. We will consider an example for this in the next chapter. In quantum optics, using dipole coupling usually results in slightly better results.

5.4 Selection rules for atoms

One of the most important questions in atomic physics and quantum optics is which levels are coupled by a given electromagnetic field. To examine this we consider a single atom whose nucleus rests at the origin ($\mathbf{R}_m = 0$, it is a very good approximation to neglect the center-of-mass motion in the derivation of selection rules). Since all the other particles are electrons, the electric dipole moment can be written as

$$\begin{aligned} \mathbf{d}_m &= \sum_{\alpha_m} q_{\alpha_m} (\mathbf{x}_{\alpha_m} - \mathbf{R}_m) \\ &= -e \sum_{\alpha_m} \mathbf{x}_{\alpha_m} \\ &=: -e\mathbf{r} \end{aligned} \quad (5.67)$$

\mathbf{r} is the sum over the position of the electrons relative to the nucleus. in the following we will omit the index m , since we consider a single atom only, $\mathbf{d}_m \rightarrow$

\mathbf{d} , $\alpha_m \rightarrow \alpha$ etc. Here, the sum includes only the electrons. If there are any contributions from the nucleus, they will be added explicitly.

We have seen in chapter 4 that atomic states are, to a very good approximation, eigenstates of the total angular momentum

$$\mathbf{F} = \mathbf{s}_K + \sum_{\alpha} (\mathbf{L}_{\alpha} + \mathbf{s}_{\alpha}) , \quad (5.68)$$

with the atomic state $|\psi\rangle = |F, m_F, \dots\rangle$. Since the interaction is mediated through the dipole moment, the transition amplitudes between $|F, m_F\rangle$ and $|F', m'_F\rangle$ are proportional to

$$\mathbf{d}_{F, m_F; F', m'_F} := \langle F, m_F | -e\mathbf{r} | F', m'_F \rangle . \quad (5.69)$$

The derivation of these matrix elements is surprisingly simple and elegant if one employs the concept of *Tensor operators*, which is summarized in appendix 10.2. In particular, a Tensor operator of order 1 is also called a *Vector operator* R_q ($q = -1, 0, 1$). In the notation of App. 10.2, the spherical vector components R_q are given by $R_q \equiv T_{1,q}$. These operators are defined by their commutator relations with a given angular momentum operator \mathbf{L} :

$$\begin{aligned} [L_+, R_+] &= 0 \\ [L_-, R_-] &= 0 \\ [L_+, R_-] &= \sqrt{2}\hbar R_0 \\ [L_-, R_+] &= \sqrt{2}\hbar R_0 \\ [L_z, R_{\pm}] &= \pm\hbar R_{\pm} \\ [L_z, R_0] &= 0 \\ [L_{\pm}, R_0] &= \sqrt{2}\hbar R_{\pm} \end{aligned} \quad (5.70)$$

Here we have defined $L_{\pm} = L_x \pm iL_y$. Equivalent to these relations is

$$[L_a, \bar{R}_b] = i\hbar \varepsilon_{abc} \bar{R}_c \quad (5.71)$$

if the Cartesian components \bar{R}_c are related to the spherical components by $\bar{R}_x = (R_- - R_+)/\sqrt{2}$ and $\bar{R}_y = i(R_- + R_+)/\sqrt{2}$ as well as $\bar{R}_z = R_0$. An example for a vector operator is the position operator of a Schrödinger particle, with \mathbf{L} the orbital angular momentum operator. However, it is an easy task to prove that the

dipole moment \mathbf{d} is a vector operator, too:

$$\begin{aligned}
 [\mathbf{F}_a, \mathbf{d}_b] &= [(\mathbf{s}_K)_a + \sum_{\alpha} (\mathbf{L}_{\alpha} + \mathbf{s}_{\alpha})_a, -e \sum_{\beta} (\mathbf{x}_{\beta})_b] \\
 &= -e \sum_{\alpha, \beta} [(\mathbf{L}_{\alpha})_a, (\mathbf{x}_{\beta})_b] \\
 &= -e \sum_{\alpha, \beta} \delta_{\alpha\beta} i\hbar \varepsilon_{abc} (\mathbf{x}_{\beta})_c \\
 &= -ie\hbar \varepsilon_{abc} \sum_{\alpha} (\mathbf{x}_{\alpha})_c \\
 &= i\hbar \varepsilon_{abc} \mathbf{d}_c
 \end{aligned} \tag{5.72}$$

To a large degree, the matrix elements of vector operators can be calculated algebraically.

$$\begin{aligned}
 \pm \hbar \langle F, m_F | \mathbf{d}_{\pm} | F', m'_F \rangle &= \langle F, m_F | [\mathbf{F}_z, \mathbf{d}_{\pm}] | F', m'_F \rangle \\
 &= \langle F, m_F | \mathbf{F}_z \mathbf{d}_{\pm} - \mathbf{d}_{\pm} \mathbf{F}_z | F', m'_F \rangle \\
 &= \hbar(m_F - m'_F) \langle F, m_F | \mathbf{d}_{\pm} | F', m'_F \rangle
 \end{aligned} \tag{5.73}$$

It follows that $\langle F, m_F | \mathbf{d}_{\pm} | F', m'_F \rangle$ can only be non-zero if $\Delta m := m_F - m'_F = \pm 1$. Since \mathbf{d}_z commutes with \mathbf{F}_z one can show that

$$\begin{aligned}
 0 &= \langle F, m_F | [\mathbf{F}_z, \mathbf{d}_z] | F', m'_F \rangle \\
 &= \hbar(m_F - m'_F) \langle F, m_F | \mathbf{d}_z | F', m'_F \rangle .
 \end{aligned} \tag{5.74}$$

In summary, we obtain the *selection rules for the magnetic quantum number*

$$\langle F, m_F | \mathbf{d}_z | F', m'_F \rangle \neq 0 \quad \text{only if} \quad m_F = m'_F \tag{5.75}$$

$$\langle F, m_F | \mathbf{d}_+ | F', m'_F \rangle \neq 0 \quad \text{only if} \quad m_F = m'_F + 1 \tag{5.76}$$

$$\langle F, m_F | \mathbf{d}_- | F', m'_F \rangle \neq 0 \quad \text{only if} \quad m_F = m'_F - 1 \tag{5.77}$$

Further statements about the matrix elements of \mathbf{d} can be derived with the aid of the

Wigner-Eckart Theorem (here only for vector operators): The spherical matrix elements

$\{\mathbf{d}_-, \mathbf{d}_0, \mathbf{d}_+\} := \{(d_x - id_y)/\sqrt{2}, \mathbf{d}_z, -(d_x + id_y)/\sqrt{2}\}$ of a vector operator are given by

$$\boxed{\langle F, m_F | \mathbf{d}_q | F', m'_F \rangle = \langle F', m'_F; 1, q | F, m_F, (1, F') \rangle \frac{\mathcal{D}(F, F')}{\sqrt{2F+1}} .} \tag{5.78}$$

The factor $\mathcal{D}(F, F')$ is called reduced matrix element.

This very important theorem states that the matrix elements of vector operators (and tensor operators in general) are proportional to Clebsch-Gordan coefficients (see Appendix 10.2 where also the proof of the theorem is presented).

The Wigner-Eckart theorem has an intuitive interpretation. Electric dipole coupling is associated with absorption or emission of a photon, which has spin one. Since the total angular momentum is conserved, one has to add the atomic and photonic angular momenta in the way described in Sec. 10.2.

The Wigner-Eckart theorem does not make any statement about the form of the reduced matrix element $\mathcal{D}(F, F')$. For a given system one can derive it by explicit calculation for a given transition m, m' . However, this is usually very difficult and demands considerable numerical efforts. One therefore either uses the experimental value or the following estimate. The dipole moment operator is defined as electron charge times electron distance from the nucleus. Hence, the reduced matrix element should be of the order of ea_0 , where a_0 is Bohr's radius. This rule of thumb gives often very reasonable estimates.

Another important question is the physical meaning of the spherical components d_q of the dipole moment. To shed light on this we enumerate the polarization vectors for left/right-circularly polarized light and linear orthogonally polarized light as

$$\epsilon_{\pm 1} = \epsilon_{\pm} = \mp \frac{1}{\sqrt{2}}(e_x \pm ie_y) \quad , \quad \epsilon_0 = e_z \quad (5.79)$$

It should be noted that these vectors are orthogonal in the sense of $\epsilon_q^* \cdot \epsilon_{q'} = \delta_{q,q'}$. With these vectors we can express the dipole vector in its spherical components,

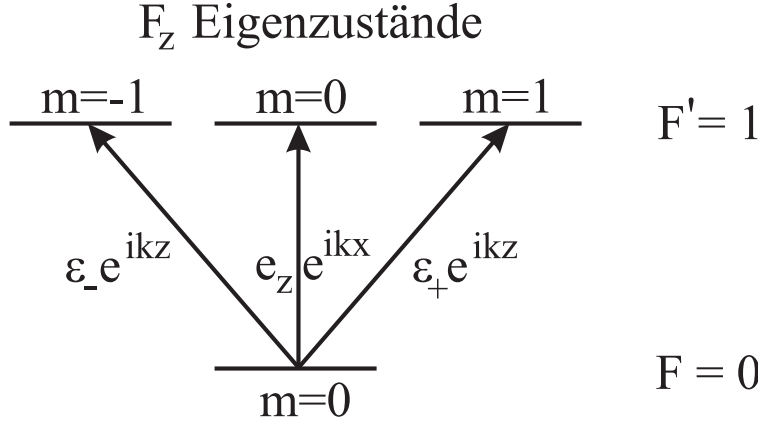
$$\mathbf{d} = d_x \mathbf{e}_x + d_y \mathbf{e}_y + d_z \mathbf{e}_z = \sum_{q=-1}^1 d_q \epsilon_q^* \quad (5.80)$$

We thus can rewrite the interaction Hamiltonian for an atom with a ground state multiplett F_g and excited state multiplett F_e by using the Wigner-Eckhart theorem,

$$\begin{aligned} H_{\text{int}} &= -\mathbf{d} \cdot \mathbf{D} \\ &= - \sum_{m_g, m_e, q} |F_e, m_e\rangle \langle F_e, m_e| d_q |F_g, m_g\rangle \langle F_g, m_g| \epsilon_q^* \cdot \mathbf{D} + \text{H.c.} \\ &= - \frac{\mathcal{D}(F_e, F_g)}{\sqrt{2F_e + 1}} \sum_{m_g, m_e, q} |F_e, m_e\rangle \langle F_g, m_g| \langle F_g, m_g; 1, q | F_e, m_e, (1, F_g) \rangle \epsilon_q^* \cdot \mathbf{D} + \text{H.c.} \end{aligned}$$

Hence, left-circularly polarized light of a laser beam running along the quantization axis (= z-axis), i.e., $\mathbf{D} = \epsilon_+ \exp(ikz) \exp(-i\omega t)$, couples to d_+ and therefore

requires $m_e = m_g + 1$. Likewise, $\boldsymbol{\varepsilon} = \mathbf{e}_z$ couples to d_0 and implies $m_e = m_g$. The corresponding laser beam needs to propagate perpendicular to the quantization axis, since its wave vector is orthogonal to \mathbf{e}_z .⁴



We have to emphasize that circularly polarized light only implies $m_e = m_g \pm 1$ if the laser beam propagates along the quantization axis. For another direction the laser light can generally couple to all spherical components d_q . For instance, a left-circularly polarized beam along the x-axis has polarization $\boldsymbol{\varepsilon} = (-\mathbf{e}_y - i\mathbf{e}_z)/\sqrt{2}$. Because $\mathbf{d} \cdot \boldsymbol{\varepsilon} = -id_0/\sqrt{2} - i(d_+ + d_-)/2$ it couples to all spherical components, so that $m_e = m_g + i$, $i = -1, 0, 1$ are possible.

To conclude this section we summarize the general dipole selection rules, which can be derived from the properties of the Clebsch-Gordan coefficients:

$$\Delta m = 0, \pm 1$$

$$\Delta J = 0, \pm 1$$

For $J' = J$ the transition $m = 0 \rightarrow m' = 0$ is forbidden.

⁴Strictly speaking, this is too strong a statement. Since real laser beams are always focused, they correspond to a wavepacket to which also wave vectors contribute which are not exactly parallel to the axis of propagation. A real laser beam therefore always has a small longitudinally polarized part. This part, however, can usually be neglected.

Chapter 6

The two-level model for atoms

If atoms in their ground state $|g\rangle$ with energy E_g are exposed to a laser beam composed of photons with energy $\hbar\omega$, they can absorb the energy of the photons and make a transition to an excited state $|e\rangle$ with energy E_e . Energy conservation implies $E_e \approx E_g + \hbar\omega$. If no other atomic energy levels are close to $E_g + \hbar\omega$, one can neglect all states but $|g\rangle$ and $|e\rangle$. One then can think of the atom as a neutral

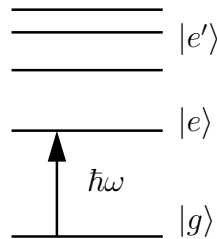
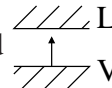


Figure 6.1: Absorbtion of a photon by an atom

particle with just two internal levels $|g\rangle$ and $|e\rangle$; it then resembles for instance a neutron with spin $\frac{1}{2}$. Models with two levels appear frequently in quantum mechanics, for instance for

- real spin $\frac{1}{2}$ particles
- photons with polarization

- the band edges of valence band and conductor band



- atoms and molecules
- ions

- Josephson effect in superconductors
- interferometer
- qbits

The mathematical content of this chapter can also be applied to the other physical systems, but we will focus on atoms and molecules.

Two-level models provide an intuitive picture of complex physical phenomena. Instead of their simplicity they often lead also to excellent quantitative results.

6.1 Derivation of two-level systems

Example: hydrogen atom The eigenstates (without spin) have the form $|nlm\rangle$. In absence of a magnetic field they are degenerated with respect to m .

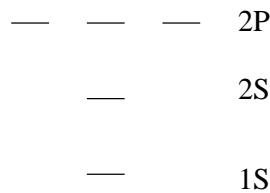
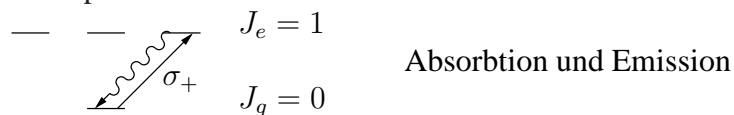


Figure 6.2: Sketch of hydrogen energy levels

If laser light of frequency $\omega \approx (E_{2P} - E_{1S})/\hbar$ is used, all multiplets except for $1S$ and $2P$ can be neglected. We then obtain a system with four states. Sometimes systems consisting of two multiplets are already called 2-level systems, since only two energy levels are involved. In the presence of hyperfine degeneracy such systems can include a considerable number of states. To achieve a real two-level system (with two *states* instead of just two energy levels), one has to use laser with a suitable polarisation and to keep an eye on spontaneous emission.

Examples for two-level systems

1. Example:



If one pumps with circularly polarized light, only the two states to the right may form a two-level system.

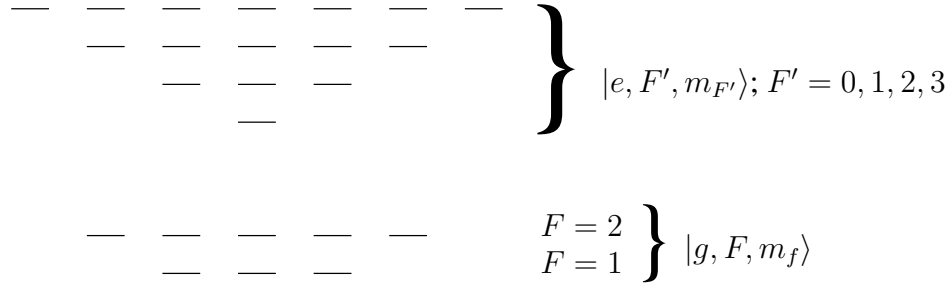
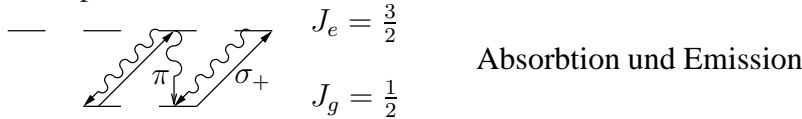


Figure 6.3: Hyperfine splitting of Rb^{87} . Even if light is used that is resonant with two energy levels (for instance $F = 1 \rightarrow F' = 2$), one generally has to consider many states.

2. Example:



Mechanism:

- $|m_g = \frac{1}{2}\rangle$ is only coupled to $|m_e = \frac{3}{2}\rangle$ and forms a two-level system
- $|m_g = -\frac{1}{2}\rangle$ is coupled to $|m_e = \frac{1}{2}\rangle$. This state decays spontaneously into $|m_g = -\frac{1}{2}\rangle$ and $|m_g = \frac{1}{2}\rangle$. Hence, for each spontaneous emission population is transferred to the real two-level system $|m_g = \frac{1}{2}\rangle$, $|m_e = \frac{3}{2}\rangle$.
- After many spontaneous emissions almost all atoms are transferred to this system.

6.1.1 Representation of operators

In the rest of this chapter we will only consider a two-level system with states $|g\rangle$, $|e\rangle$. The representation of any operator acting on this system can be found as follows:

$$\hat{O} = \sum_{n=0}^{\infty} |n\rangle \langle n| \hat{O} \sum_{m=0}^{\infty} |m\rangle \langle m| \approx \sum_{n,m=e,g} |n\rangle O_{nm} \langle m|$$

$$\text{mit: } O_{nm} = \langle n | \hat{O} | m \rangle = \begin{pmatrix} O_{ee} & O_{eg} \\ O_{ge} & O_{gg} \end{pmatrix}$$

The most important operators are:

Identity operator $\mathbb{1} = \begin{pmatrix} 1 & \\ & 1 \end{pmatrix}$

Hamiltonian $H_0 = \begin{pmatrix} E_e & \\ & E_g \end{pmatrix}$ since $|e\rangle, |g\rangle$ are eigenstates of H_0 .

Electronic position and dipole operator $\mathbf{x} \propto \mathbf{d} = -e\mathbf{x}$ with matrix elements

$$\begin{aligned}\langle e|\mathbf{d}|g\rangle &:= \mathbf{d}_{eg} = \mathbf{d}_{ge}^* \\ \langle e|\mathbf{d}|e\rangle &= \langle g|\mathbf{d}|g\rangle = 0\end{aligned}$$

The vanishing of the diagonal elements is a consequence of $|e\rangle$ and $|g\rangle$ being eigenstates of the parity operator, and of \mathbf{d} having parity -1 . One therefore has

$$\mathbf{d} = \mathbf{d}_{eg} \cdot \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} + \mathbf{d}_{eg}^* \cdot \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} = \begin{pmatrix} 0 & \mathbf{d}_{eg} \\ \mathbf{d}_{eg}^* & 0 \end{pmatrix} = -e \begin{pmatrix} 0 & \mathbf{x}_{eg} \\ \mathbf{x}_{eg}^* & 0 \end{pmatrix} \quad (6.1)$$

where \mathbf{x}_{eg} is the matrix element of the relative coordinate of the electrons.

Electronic momentum operator We first proceed as with the position operator to find $\langle g|\mathbf{p}|g\rangle = 0$ and $\langle e|\mathbf{p}|e\rangle = 0$. To derive the off-diagonal elements we use the following trick:

$$[\mathbf{x}, H_0] = \frac{i\hbar}{m_e} \mathbf{p} \quad (6.2)$$

This follows from $H_0 = \sum_{\alpha} \frac{1}{2m_e} \mathbf{p}_{\alpha}^2 + \sum_{\alpha, \beta} V(\alpha, \beta)$ and $\mathbf{p} = \sum_{\alpha} \mathbf{p}_{\alpha}$, $\mathbf{x} = \sum_{\alpha} \mathbf{x}_{\alpha}$:

$$\begin{aligned}[\mathbf{x}_{\alpha}, H_0] &= \sum_{\alpha'} \frac{1}{2m_e} [\mathbf{x}_{\alpha}, \mathbf{p}_{\alpha'}^2] \\ &= \frac{i\hbar}{m_e} \mathbf{p}_{\alpha}\end{aligned}$$

Hence

$$\begin{aligned}\langle e|\mathbf{p}|g\rangle &= \frac{m_e}{i\hbar} \langle e|[\mathbf{x}, H_0]|g\rangle \\ &= \frac{im_e}{\hbar} (E_e - E_g) \langle e|\mathbf{x}|g\rangle\end{aligned}$$

so that we find for the matrix representation of the electronic momentum operator

$$\mathbf{p} = \frac{m_e \omega_0}{e} \begin{pmatrix} 0 & -i\mathbf{d}_{eg} \\ i\mathbf{d}_{eg}^* & 0 \end{pmatrix} \quad (6.3)$$

$$\omega_0 = \frac{E_e - E_g}{\hbar}. \quad (6.4)$$

6.1.2 Coupling between a two-level atom and the electromagnetic field

Consider an electromagnetic wave with a positive frequency part of the vector potential of the form

$$\mathbf{A}^{(+)}(t, \mathbf{R} = 0) = \mathbf{A}_0 e^{-i\omega_L t} \quad \mathbf{R}: \text{Center-of-mass of the atom} \quad (6.5)$$

The interaction Hamiltonian in dipole approximation reads ($\mathbf{E} = -\dot{\mathbf{A}}$)

$$\begin{aligned} H_{int} &= -\mathbf{d} \cdot \mathbf{E} \\ &= -\mathbf{d}(\mathbf{E}^{(+)} + \mathbf{E}^{(-)}) \\ &= -i\omega_L (\mathbf{d}_{eg} |e\rangle \langle g| + \mathbf{d}_{eg}^* |g\rangle \langle e|) (\mathbf{A}_0 e^{-i\omega_L t} - \mathbf{A}_0^* e^{i\omega_L t}) \end{aligned} \quad (6.6)$$

For comparison we present the minimal coupling Hamiltonian:

$$H'_{int} = \frac{e}{m_e} \mathbf{p} \cdot \mathbf{A} = -i\omega_0 (\mathbf{d}_{eg} |e\rangle \langle g| - \mathbf{d}_{eg}^* |g\rangle \langle e|) (\mathbf{A}_0 e^{-i\omega_L t} + \mathbf{A}_0^* e^{i\omega_L t}) \quad (6.7)$$

Apart from various signs, which can be transformed away by a different choice of the phase of the basis vectors and a shift in the center-of-mass coordinate, the main difference is the occurrence of the factors ω_L and ω_0 , respectively. The reason for this difference is that we only consider a part of the full Hamiltonian (that includes all levels), and only the full Hamiltonians are unitarily equivalent. In Fig. 6.4 the unitary transformation is represented as a rotation in Hilbert space. This has the

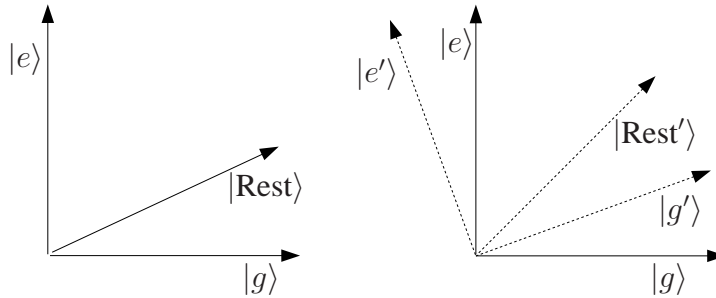


Figure 6.4: Unitary transformation $|\psi'\rangle = U |\psi\rangle$

following meaning: the unitary Power-Zieman-Woolley transformation “rotates” the states in Hilbert space. The two-level approximation is a projection onto a two-dimensional subspace. Depending on whether one performs this projection before ($|e\rangle$ and $|g\rangle$) or after ($|e'\rangle$ and $|g'\rangle$) the Power-Zieman-Woolley transformation, one

obtains different results since one considers different subspaces.

In practical calculations this difference is unimportant since we are only interested in the case $\omega_0 \approx \omega_L$.¹ In the following we will use the $-\mathbf{d} \cdot \mathbf{E}$ coupling. We define² *Rabi frequency*

$$\Omega := \frac{1}{\hbar} \mathbf{d}_{eg} \cdot \mathbf{E}_0 = \frac{1}{\hbar} \mathbf{d}_{eg} \cdot \mathbf{A}_0 \omega_L \quad (6.8)$$

with $\mathbf{E}_0 := \mathbf{A}_0 \omega_L$ and the less important abbreviation

$$\tilde{\Omega} = \frac{1}{\hbar} \mathbf{d}_{eg} \mathbf{E}_0^* \quad (6.9)$$

We arrive at

$$H_{int} = -i\hbar |e\rangle \langle g| (\Omega e^{-i\omega_L t} - \tilde{\Omega} e^{i\omega_L t}) + i\hbar |g\rangle \langle e| (\Omega^* e^{i\omega_L t} - \tilde{\Omega}^* e^{-i\omega_L t}) \quad (6.10)$$

The unperturbed Hamiltonian is given by

$$H_0 = \begin{pmatrix} E_e & 0 \\ 0 & E_g \end{pmatrix} = \frac{1}{2}(E_e + E_g)\mathbb{1} + \frac{1}{2}(E_e - E_g)\sigma_z \quad (6.11)$$

By choosing a particular value for E_g (the zero-point of energy can be chosen at will) one can represent H_0 in various ways:

$$E_e + E_g = 0 \Rightarrow H_0 = \frac{\hbar\omega_0}{2}\sigma_z, \quad \omega_0 = \frac{E_e - E_g}{\hbar} \quad (6.12)$$

$$E_g = 0 \Rightarrow H_0 = (E_e - E_g) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} = \hbar\omega_0 \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \quad (6.13)$$

We will frequently make use of the *rotating-wave approximation*. This is a widely used and well-justified approximation which considerably simplifies calculations. To derive it, we first switch to the interaction picture. Thereby the temporal evolution associated with H_0 is already incorporated in the wave function:

$$|\psi\rangle = e^{-iH_0 t/\hbar} |\tilde{\psi}\rangle = U |\tilde{\psi}\rangle \quad (6.14)$$

The transformed Schrödinger equation becomes $i\hbar |\dot{\tilde{\psi}}\rangle = \tilde{H} |\tilde{\psi}\rangle$ and the Hamiltonian is transformed according to

$$\begin{aligned} \tilde{H} &= -i\hbar U^\dagger \dot{U} + U^\dagger (H_0 + H_{int}) U = U^\dagger H_{int} U \\ &= -i\hbar |e\rangle \langle g| (\Omega e^{-i(\omega_L - \omega_0)t} - \tilde{\Omega} e^{i(\omega_L + \omega_0)t}) + \text{H.c.} \end{aligned} \quad (6.15)$$

¹ $\omega_0 \approx \omega_L$ is a requirement for the two-level model to be valid.

²Very often the definitions used in the literature differ from our convention by a factor of two.

The term $e^{i(\omega_L + \omega_0)t}$ is fastly oscillating and therefore averages to zero during the evolution. This leads to the following simple form of the Hamiltonian in interaction picture,

$$\boxed{\tilde{H}_{WW}(t) = -i\hbar e^{-i\Delta t} \Omega |e\rangle \langle g| + i\hbar \Omega^* e^{i\Delta t} |g\rangle \langle e|} \quad (6.16)$$

where we have used the laser's detuning $\Delta = \omega_L - \omega_0$.

Transforming back to the Schrödinger picture we find

$$\boxed{H = \frac{\hbar\omega_0}{2}\sigma_z + \hbar \begin{pmatrix} 0 & -i\Omega e^{-i\omega_L t} \\ i\Omega^* e^{i\omega_L t} & 0 \end{pmatrix}} \quad (6.17)$$

By another unitary transformation we can achieve that H only depends on the absolute value of Ω . This transformation is achieved by multiplying $|e\rangle$ with a phase factor $e^{i\varphi}$, where $-i\Omega = |\Omega|e^{i\varphi}$.

$$\begin{aligned} U : |e\rangle &\rightarrow e^{i\varphi} |e\rangle \\ U &= \begin{pmatrix} e^{i\varphi} & 0 \\ 0 & 1 \end{pmatrix} \end{aligned} \quad (6.18)$$

This leads to the final and simple form for the Hamiltonian,

$$U^\dagger H U = \frac{\hbar\omega_0}{2}\sigma_z + \hbar \begin{pmatrix} 0 & |\Omega|e^{-i\omega_L t} \\ |\Omega|e^{i\omega_L t} & 0 \end{pmatrix} \quad (6.19)$$

6.2 Rabi oscillations and Landau-Zener transitions

We will now deal with the time evolution of a state with Hamiltonian H as discussed in the previous section.

$$i\hbar\partial_t \begin{pmatrix} \psi_e \\ \psi_g \end{pmatrix} = \hbar \begin{pmatrix} \frac{\omega_0}{2} & -i\Omega e^{-i\omega_L t} \\ i\Omega^* e^{i\omega_L t} & -\frac{\omega_0}{2} \end{pmatrix} \begin{pmatrix} \psi_e \\ \psi_g \end{pmatrix} \quad (6.20)$$

Without the time dependent factor $e^{i\omega_L t}$ this equation would be easily solvable. Luckily, we can remove this factor by a unitary transformation:

$$U = e^{-i\frac{\omega_L}{2}\sigma_z t} = \begin{pmatrix} e^{-i\frac{\omega_L}{2}t} & 0 \\ 0 & e^{i\frac{\omega_L}{2}t} \end{pmatrix} \quad \begin{pmatrix} \psi_e \\ \psi_g \end{pmatrix} = U \begin{pmatrix} \tilde{\psi}_e \\ \tilde{\psi}_g \end{pmatrix} \quad (6.21)$$

The transformed equation becomes

$$\begin{aligned} i\hbar\partial_t \begin{pmatrix} \tilde{\psi}_e \\ \tilde{\psi}_g \end{pmatrix} &= \left(-i\hbar U^\dagger \dot{U} + U^\dagger H U \right) \begin{pmatrix} \tilde{\psi}_e \\ \tilde{\psi}_g \end{pmatrix} \\ &= \left\{ -\frac{\hbar\omega_L}{2}\sigma_z + \frac{\hbar\omega_0}{2}\sigma_z + \hbar \begin{pmatrix} 0 & -i\Omega \\ i\Omega^* & 0 \end{pmatrix} \right\} \begin{pmatrix} \tilde{\psi}_e \\ \tilde{\psi}_g \end{pmatrix} \end{aligned} \quad (6.22)$$

We therefore find for \tilde{H} :

$$\boxed{\tilde{H} = \hbar \begin{pmatrix} -\frac{\Delta}{2} & -i\Omega \\ i\Omega^* & \frac{\Delta}{2} \end{pmatrix}} \quad (6.23)$$

Such a time-independent Hamiltonian is only possible in rotating-wave approximation. The counter-rotating terms would still depend on time after transformation Eq. (6.21). We now can easily solve the Schrödinger equation:

$$\begin{pmatrix} \tilde{\psi}_e(t) \\ \tilde{\psi}_g(t) \end{pmatrix} = e^{-it\tilde{H}/\hbar} \begin{pmatrix} \tilde{\psi}_e(0) \\ \tilde{\psi}_g(0) \end{pmatrix} \quad (6.24)$$

Let us now rewrite \tilde{H} . Since the Hamiltonian is Hermitean and traceless, there are only three independent parameters.

$$\tilde{H} = \hbar \mathbf{n} \cdot \boldsymbol{\sigma}, \quad \mathbf{n} = \begin{pmatrix} \text{Im}\Omega \\ \text{Re}\Omega \\ -\frac{\Delta}{2} \end{pmatrix} \quad (6.25)$$

The expression $e^{-it\mathbf{n} \cdot \boldsymbol{\sigma}}$ can be expanded into a Taylor series,

$$e^{-it\mathbf{n} \cdot \boldsymbol{\sigma}} = \sum_{l=0}^{\infty} \frac{1}{l!} (-it)^l (\mathbf{n} \cdot \boldsymbol{\sigma})^l \quad (6.26)$$

The product of two σ matrices is given by

$$\sigma_i \sigma_j = i\varepsilon_{ijk} \sigma_k + \delta_{ij} \mathbb{1} \quad (6.27)$$

$$\Rightarrow (\mathbf{n} \cdot \boldsymbol{\sigma})^2 = n_i \sigma_i n_j \sigma_j = n_i n_j (i\varepsilon_{ijk} \sigma_k + \delta_{ij} \mathbb{1}) = |\mathbf{n}|^2 \mathbb{1} \quad (6.28)$$

$$(\mathbf{n} \cdot \boldsymbol{\sigma})^{2l} = |\mathbf{n}|^{2l} \mathbb{1} \quad (6.29)$$

We thus can bring $e^{-it\mathbf{n} \cdot \boldsymbol{\sigma}}$ into the form

$$e^{-it\mathbf{n} \cdot \boldsymbol{\sigma}} = \sum_{l=0}^{\infty} \frac{(-it)^{2l}}{(2l)!} (\mathbf{n} \cdot \boldsymbol{\sigma})^{2l} + \sum_{l=0}^{\infty} \frac{(-it)^{2l+1}}{(2l+1)!} (\mathbf{n} \cdot \boldsymbol{\sigma})^{2l+1} \quad (6.30)$$

$$\boxed{e^{-it\mathbf{n} \cdot \boldsymbol{\sigma}} = \cos(|\mathbf{n}|t) \mathbb{1} - i \frac{\mathbf{n} \cdot \boldsymbol{\sigma}}{|\mathbf{n}|} \sin(|\mathbf{n}|t)} \quad (6.31)$$

We introduce the notation

$$w \equiv |\mathbf{n}| = \sqrt{\left(\frac{\Delta}{2}\right)^2 + |\Omega|^2} \quad (6.32)$$

which only depends on the modulus of Ω . For an atom that initially is in its ground state, the solution becomes

$$\begin{pmatrix} \tilde{\psi}_e(0) \\ \tilde{\psi}_g(0) \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \Rightarrow \boxed{\begin{pmatrix} \tilde{\psi}_e(t) \\ \tilde{\psi}_g(t) \end{pmatrix} = \begin{pmatrix} -\frac{\Omega}{w} \sin(wt) \\ \cos(wt) - i\frac{\Delta}{2w} \sin(wt) \end{pmatrix}} \quad (6.33)$$

The probability amplitude $\tilde{\psi}_e(t)$ to excite an atom oscillates with frequency w , this phenomenon is called *Rabi oscillations*. We have

$$\mathcal{P}_e(t) = |\tilde{\psi}_e(t)|^2 = \frac{|\Omega|^2}{w^2} \sin^2(wt) \quad (6.34)$$

The maximum value of \mathcal{P}_e is reached for $wt = \pi/2 + n\pi$ and given by

$$\mathcal{P}_e \propto \frac{|\Omega|^2}{\left(\frac{\Delta}{2}\right)^2 + |\Omega|^2} \quad (6.35)$$

If the laser is not detuned against the resonance frequency, $\Delta = 0$, complete excitation ($\mathcal{P}_e = 1$) is possible. \mathcal{P}_e depends strongly on the detuning,

$$\Delta \gg |\Omega| \Rightarrow \mathcal{P}_e \approx \frac{|\Omega|^2}{\Delta^2} \ll 1 \quad (6.36)$$

The excitation probability therefore depends on the ratio of Rabi frequency and detuning.³ These results are of very high relevance for atom optics, quantum optics, electron spin resonance, nuclear spin resonance, Interferometry, and many other areas.

With laser pulses of a particular duration one can prepare well-defined atomic states. The so-called ideal $\frac{\pi}{2}$ pulse⁴ creates a 50-50 superposition of ground and excited state:

$$\Delta = 0 \quad t = \frac{\pi}{4\Omega} \Rightarrow U = e^{-it\mathbf{n}\cdot\boldsymbol{\sigma}} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix} \quad (6.37)$$

The ideal π pulse transfers all electrons to the excited state if they were prepared in the ground state, or vice versa:

$$\Delta = 0 \quad t = \frac{\pi}{2\Omega} \Rightarrow U = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \quad (6.38)$$

³It is often claimed that the excitation probability depends on the ratio of Rabi frequency and spontaneous emission rate. We will see later that this ratio is also important for small detunings. The ratio of Ω/Δ is usually more important, though.

⁴In practise the detuning of a laser against the resonance frequency of an atom depends through the Doppler effect also on its center-of-mass motion. At room temperature this makes an ideal $\pi/2$ pulse impossible. For cold atoms and with some tricks one can achieve such transitions with very good efficiency.

To visualize the states of a two-level system one can use the Bloch vector, whose components are given by $\langle \sigma \rangle = (v, w, u)$. One then has

$$\begin{aligned} u &= \langle \sigma_z \rangle = |\psi_e|^2 - |\psi_g|^2 \\ v &= \langle \sigma_x \rangle = \psi_e^* \psi_g + \psi_g^* \psi_e \\ w &= \langle \sigma_y \rangle = -i(\psi_e^* \psi_g - \psi_g^* \psi_e) \end{aligned} \quad (6.39)$$

The Bloch vector of a pure state is of unit length, it always moves on the Bloch sphere of radius 1 around the origin.

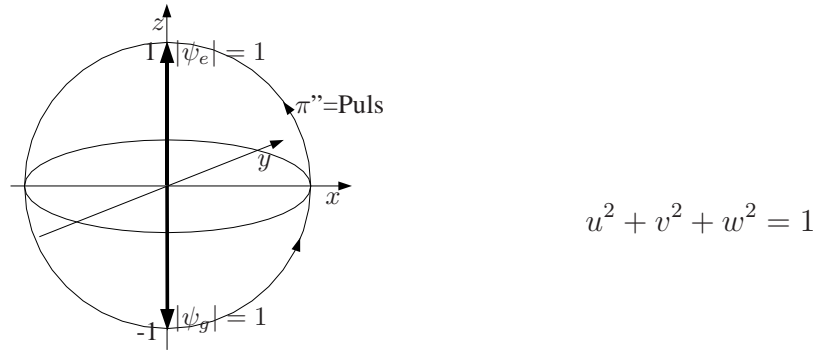


Figure 6.5: The Bloch sphere

For a non-detuned laser beam Rabi oscillations have the following form,

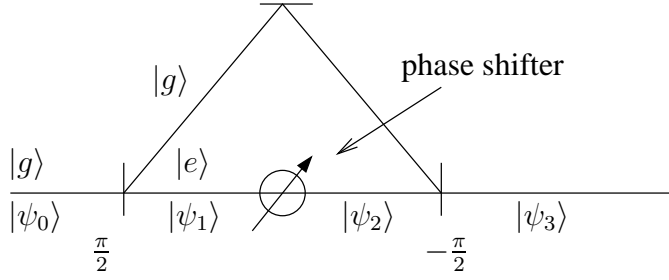
$$\begin{pmatrix} \psi_e \\ \psi_g \end{pmatrix} = \begin{pmatrix} -\sin(\Omega t) \\ \cos(\Omega t) \end{pmatrix} \quad (6.40)$$

The corresponding Bloch vector becomes

$$\begin{pmatrix} v \\ w \\ u \end{pmatrix} = \begin{pmatrix} -\sin(2\Omega t) \\ 0 \\ -\cos(2\Omega t) \end{pmatrix} \quad (6.41)$$

For a π pulse, $t = \frac{\pi}{2\Omega}$, the Bloch vector moves on a semi-circle, see Fig. 6.5.

Interferometry with two internal states: an example for the application of π and $\pi/2$ pulses two-level systems and Rabi oscillations can be used to realize an interferometer. The two states thereby play the role of the arms of the interferometer, Rabi oscillations provide beam splitters and mirrors. Sketch of set-up:



$$|\psi_0\rangle = |g\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

$$\begin{aligned} |\psi_1\rangle &= U_{\pi/2} |\psi_0\rangle \\ &= \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} \\ &= \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ 1 \end{pmatrix} \\ &\Leftrightarrow \frac{1}{\sqrt{2}} (|g\rangle - |e\rangle) \end{aligned}$$

$$\begin{aligned} |\psi_2\rangle &= \begin{pmatrix} e^{i\varphi} & 0 \\ 0 & 1 \end{pmatrix} |\psi_1\rangle \\ &= \frac{1}{\sqrt{2}} (|g\rangle - e^{i\varphi} |e\rangle) \end{aligned}$$

$$\begin{aligned} |\psi_3\rangle &= U_{-\pi/2} |\psi_2\rangle \\ &= \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix} \frac{1}{\sqrt{2}} \begin{pmatrix} -e^{i\varphi} \\ 1 \end{pmatrix} \\ &= \frac{1}{2} \begin{pmatrix} 1 - e^{-i\varphi} \\ 1 + e^{i\varphi} \end{pmatrix} \\ &\Leftrightarrow \frac{1}{2} (|g\rangle (1 + e^{i\varphi}) + |e\rangle (1 - e^{i\varphi})) \end{aligned}$$

Detect the number of atoms in $|e\rangle$:

$$\begin{aligned} P_e &= |\langle \psi_3 | e \rangle|^2 \\ &= \left| \frac{1}{2} (1 - e^{i\varphi}) \right|^2 = \frac{1}{2} (1 - \cos \varphi) \end{aligned}$$

6.2.1 Landau-Zener transitions

Landau-Zener transitions are not as important as Rabi oscillations, but they nevertheless explain many relevant phenomena. The difference to Rabi oscillations is that the detuning is allowed to vary in time, $\Delta = \Delta(t) = \dot{\Delta}_0 t$

$$H = \hbar \begin{pmatrix} -\frac{\dot{\Delta}_0}{2}t & \Omega \\ \Omega & \frac{\dot{\Delta}_0}{2}t \end{pmatrix}$$

where we use the rotating-wave approximation and assume $\Omega \in \mathbb{R}$ for simplicity.

Equations of motion:

$$\begin{aligned} i\dot{\psi}_e &= -\frac{1}{2}\dot{\Delta}_0 t \psi_e + \Omega \psi_g \\ i\dot{\psi}_g &= \frac{1}{2}\dot{\Delta}_0 t \psi_g + \Omega \psi_e \end{aligned} \quad (6.42)$$

Differentiate $\dot{\psi}_g$ with respect to time to obtain

$$i\ddot{\psi}_g = \frac{1}{2}\dot{\Delta}_0 \psi_g + \frac{1}{2}\dot{\Delta}_0 t \dot{\psi}_g + \Omega \dot{\psi}_e$$

Inserting (6.42) results in

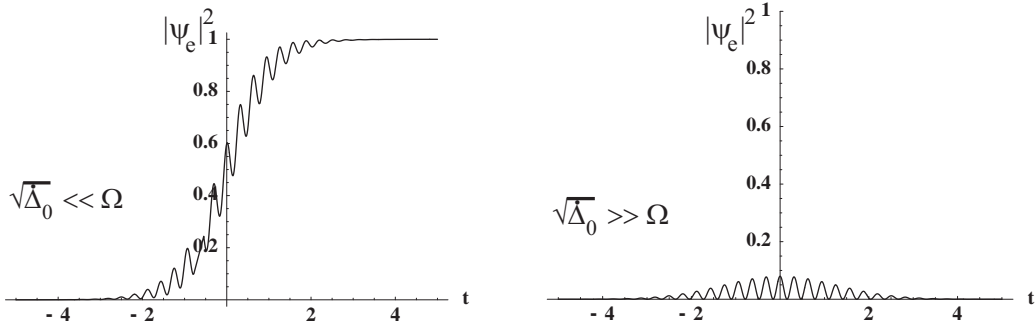
$$\ddot{\psi}_g = -\left(\Omega^2 + \frac{t^2 \dot{\Delta}_0^2}{4} + \frac{i}{2}\dot{\Delta}_0\right) \psi_g$$

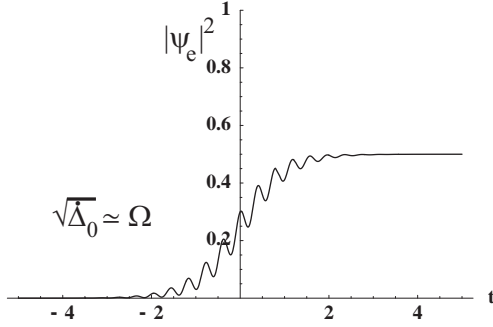
Introduce the new variable $y \equiv t\sqrt{\dot{\Delta}_0}$

$$\Rightarrow \psi_g'' = -\left(\frac{y^2}{4} + \frac{\Omega^2}{\dot{\Delta}_0} + \frac{i}{2}\right) \psi_g$$

This is the standard form of the differential equation for parabolic cylinder functions (see [17]).

The solution can be expressed in terms of confluent hypergeometric functions and, for $\psi_g(-\infty) = 1$, typically look like this:

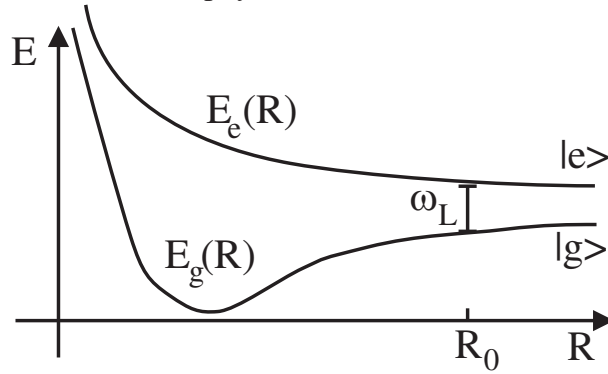




The asymptotic excitation probability $P_0(+\infty)$ can be calculated analytically and is given by

$$P_0(+\infty) = 1 - |\psi_g(+\infty)|^2 = 1 - e^{-\pi \frac{\Omega^2}{2\Delta_0}}$$

Landau-Zener transitions are of importance in many different areas. They have first been discussed in molecular physics:



- Light of frequency ω_L is only resonant for a particular distance R_0 between the nuclei.
- The time evolution of the electrons is described by states $|e(R)\rangle, |g(R)\rangle$ and is much faster than the nuclear motion (since $M_p \gg M_e$)
- In the equation of motion for the electrons, the distance between the nuclei then only enters as a (slowly varying) parameter:

$$i\hbar \begin{pmatrix} \dot{\psi}_e \\ \dot{\psi}_g \end{pmatrix} = \begin{pmatrix} -\Delta E(R(t))/2 & \Omega \\ \Omega & \Delta E(R(t))/2 \end{pmatrix} \begin{pmatrix} \psi_e \\ \psi_g \end{pmatrix}$$

- Around R_0 one has $\Delta E(R(t)) = E_e - E_g \approx \underbrace{\Delta E(R_0)}_{\hbar\omega_L} + E' \underbrace{(R - R_0)}_{\approx v_0 t}$
- $$\Rightarrow \Delta(t) = \omega_L - \Delta E(R(t))/\hbar = -E'v_0 t/\hbar$$

Landau-Zener transitions are also important for STIRAP and Bloch oscillations.

6.3 Dressed States

Dressed States are generally eigenstates of the full Hamiltonian H , including an interaction with one or few light modes. The simplest example is the interaction with a laser beam as described above,

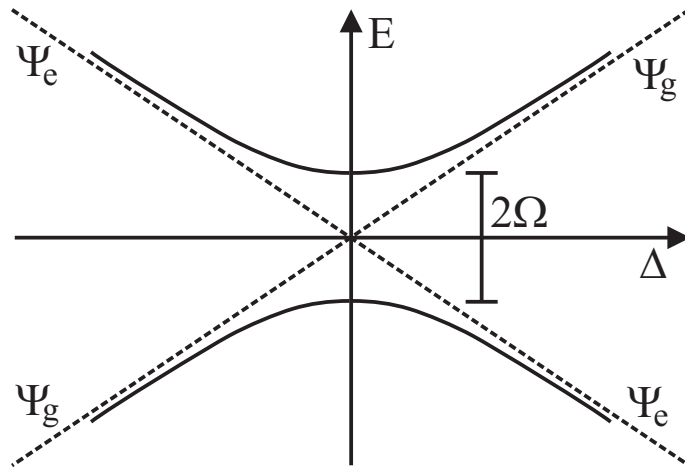
$$H\psi = E\psi$$

$$H = \hbar \begin{pmatrix} -\frac{\Delta}{2} & \Omega \\ \Omega^* & \frac{\Delta}{2} \end{pmatrix}$$

The corresponding eigenvalues are given by

$$E_{\pm} = \pm \hbar w = \pm \hbar \sqrt{\left(\frac{\Delta}{2}\right)^2 + |\Omega|^2}$$

$$\psi^{(\pm)} = \frac{1}{\sqrt{2w}} \begin{pmatrix} \frac{\Omega}{\sqrt{w \pm \Delta/2}} \\ \pm \sqrt{w \pm \Delta/2} \end{pmatrix}$$



Asymptotic behaviour for $\Delta \gg |\Omega|$. Use

$$w = \frac{|\Delta|}{2} \sqrt{1 + \frac{|\Omega|^2}{\Delta^2/4}} \approx \frac{|\Delta|}{2} \left(1 + \frac{1}{2} \frac{|\Omega|^2}{\Delta^2/4} + \dots \right)$$

$$\begin{aligned}\psi_e^{(+)} &\rightarrow \begin{cases} 0 & \Delta \rightarrow \infty \\ 1 & \Delta \rightarrow -\infty \end{cases} \\ \psi_e^{(-)} &\rightarrow \begin{cases} 1 & \Delta \rightarrow \infty \\ 0 & \Delta \rightarrow -\infty \end{cases}\end{aligned}$$

This phenomenon is called “avoided crossing”: For $\Omega = 0$ the eigenstates are ψ_e and ψ_g ; their eigenvalues are crossing at $\Delta = 0$. For $\Omega \neq 0$, the coupling of the states by the laser beam transforms this crossing into two non-intersecting hyperbolas with minimum energy distance $2\hbar |\Omega|$.

It is remarkable that, if one follows one of the hyperbolas, one evolves for E_- from ψ_g to ψ_e , and for E_+ from ψ_e to ψ_g . This means that, for time-dependent $\Delta(t)$, one can make a transition from ψ_g to ψ_e while always staying in an instantaneous eigenstate of $H(t)$ (see also the section on the adiabatic theorem below).

This behaviour is closely related to Landau-Zener transitions. tunneling between E_- and E_+ corresponds to not exciting the atoms. For a slow rate of change an atom tries to stay on one of the hyperbolas for a Landau-Zener transition.

The asymptotic transition probability can also be understood by estimating the tunneling probability between the two hyperbolas. According to the WKB approximation (see, e.g., Ref. [18]), the tunneling probability between the hyperbolas can be expressed as $P = e^{-\Delta E t / \hbar}$, with $\frac{\Delta E}{\hbar} = 2 |\Omega|$. The typical time scale t for tunneling can be estimated by the time when the distance between the two energy levels has grown to $4 |\Omega|$.

$$\begin{aligned}\Leftrightarrow 4 |\Omega|^2 &= \left(\frac{\Delta(t)}{2} \right)^2 + |\Omega|^2 \\ \Rightarrow \Delta &= \dot{\Delta}_0 t \approx 4 |\Omega| \\ \Rightarrow t &= \frac{4 |\Omega|}{\dot{\Delta}_0} \\ \Rightarrow P &= e^{-\frac{2|\Omega|4|\Omega|}{\dot{\Delta}_0}} = e^{-\frac{8|\Omega|^2}{\dot{\Delta}_0}}\end{aligned}$$

6.3.1 Dressed states in a cavity

The notion of a dressed states is more frequently used if the atoms are coupled to the quantized radiation field. The form of the corresponding two-level Hamiltonian can be derived as before. The only difference is that the Rabi frequency becomes an operator:

$$\hat{\Omega} = \frac{1}{\hbar} \mathbf{d}_{eg} \cdot \hat{\mathbf{E}}^{(+)}(\mathbf{x}) \quad (6.43)$$

This results in the Hamilton operator

$$H = \frac{\hbar\omega_0}{2}\sigma_z - \left[\mathbf{d}_{eg} \cdot \hat{\mathbf{E}}(\mathbf{x}) |e\rangle \langle g| + \mathbf{d}_{eg}^* \cdot \hat{\mathbf{E}}(\mathbf{x}) |g\rangle \langle e| \right] + H_{rad} \quad (6.44)$$

Using

$$U = e^{-i\frac{\omega_0}{2}\sigma_z t} e^{-i\frac{H_{rad}t}{\hbar}} \quad (6.45)$$

we can transform this to the interaction picture,

$$H_{int} = - \left[\mathbf{d}_{eg} |e\rangle \langle g| e^{i\omega_0 t} + \mathbf{d}_{eg}^* |g\rangle \langle e| e^{-i\omega_0 t} \right] \cdot \hat{\mathbf{E}}(\mathbf{x}, t) \quad (6.46)$$

We have

$$\hat{\mathbf{E}}(\mathbf{x}, t) = i \int d^3k \sum_{\sigma} \sqrt{\frac{\hbar\omega_k}{2(2\pi)^3\varepsilon_0}} e^{i\mathbf{k}\cdot\mathbf{x}} e^{-i\omega_k t} \boldsymbol{\epsilon}_{k\sigma} a_{k\sigma} + \text{h. c.} \quad (6.47)$$

Visualization of the terms $|e\rangle \langle g| a_{k\sigma}$ and $|e\rangle \langle g| a_{k\sigma}^\dagger$:

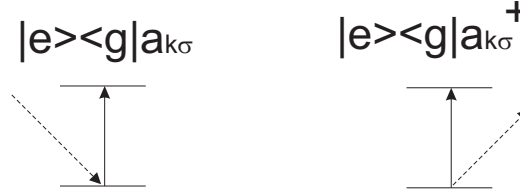


Figure 6.6: The term on the right-hand side is somehow unphysical and is neglected in the rotating-wave approximation (RWA)

In RWA, one therefore neglects the processes of *absorption & de-excitation* and *emission & excitation*.

Having made the RWA, we transform back to the Schrödinger picture,

$$H_{Schröd} = \frac{\hbar\omega_0}{2}\sigma_z + \int d^3k \sum_{\sigma} \hbar\omega_k a_{k\sigma}^\dagger a_{k\sigma} + \hbar \int d^3k \sum_{\sigma} \left[|e\rangle \langle g| a_{k\sigma} g_{k\sigma} + \text{H. c.} \right] \quad (6.48)$$

where $g_{k\sigma}$ is given by

$$g_{k\sigma} = -i\mathbf{d}_{eg} \cdot \boldsymbol{\epsilon}_{k\sigma} \sqrt{\frac{\omega_k}{2\hbar(2\pi)^3\epsilon_0}} e^{i\mathbf{k}\cdot\mathbf{x}} \quad (6.49)$$

The Jaynes-Cummings model, which describes the interaction with a single photon mode, has been particularly well studied. From Eq. (6.48) one finds

$$H = \hbar \left(\frac{\omega_0}{2} \sigma_z + \omega_L a^\dagger a + g \sigma^+ a + g^* \sigma^- a^\dagger \right) . \quad (6.50)$$

where σ^\pm are defined by

$$\sigma^+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \quad \sigma^- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \quad (6.51)$$

This model is meaningful if

- only one photon mode is occupied ($n_a \gg n_{k\sigma}$),
- only one mode of the cavity is resonant.

During the last 15 years huge progress has been made in cavity QED. The fundamental idea can be explained with the aid of an idealized cavity whose walls are perfect mirrors. The mirrors impose boundary conditions on the electrodynamic (quantum) field, which guarantee that only modes with wavelengths $\lambda_n = 2L/n$ are allowed. The corresponding frequencies $\omega_n = 2\pi c/\lambda_n \propto n$ are discrete and are only resonant for $\omega_n \approx (E_e - E_g)/\hbar$. Therefore, only modes with a certain value of n need to be taken into account. Typical values for microwaves are cavities with a length $L \approx 1$ cm and $n \approx 1$; for optical frequencies, one usually employs cavities of length $L \approx 1 - 10$ cm and $n \approx 10^6 \gg 1$.

Real cavities are never ideal. A measure for their quality is their finesse Q , which represents the mean number of round-trips which a photon makes before it is lost. Very good cavities have $Q \approx 10^6$. Because the mirror material of the cavity walls is usually slightly birefringent, a cavity is only resonant for one direction of the photon's polarization.

If an atom is placed into a 3D cavity, its spontaneous emission is modified since it can only emit photons in certain modes (of which none may be resonant), and because it can reabsorb the photons since they stay close to the atom.

Dressed states in a cavity can generally be represented by

$$|\psi\rangle = \sum_n (\psi_e(n) |e, n\rangle + \psi_g(n) |g, n\rangle) \quad (6.52)$$

We are looking for the eigenstates that obey $H|\psi\rangle = E|\psi\rangle$. This leads to the following system of equations,

$$(E + \frac{\hbar\omega_0}{2} - n\hbar\omega_L)\psi_g(n) = \hbar g^* \sqrt{n} \psi_e(n-1) \quad (6.53)$$

$$(E - \frac{\hbar\omega_0}{2} - (n-1)\hbar\omega_L)\psi_e(n-1) = \hbar g^* \sqrt{n} \psi_g(n) \quad (6.54)$$

where we choose $\psi_e(n-1)$ for the second equation because it is this component which couples to $\psi_g(n)$ (exciting the atom is accompanied by the absorption of a photon!). Setting $\Omega_n := g\sqrt{n}$ and $\Delta = \omega_L - \omega_0$ we can write this as

$$\frac{1}{\hbar} \left(E - (n - \frac{1}{2})\hbar\omega_L \right) \begin{pmatrix} \psi_e(n-1) \\ \psi_g(n) \end{pmatrix} = \begin{pmatrix} -\frac{\Delta}{2} & \Omega_n \\ \Omega_n^* & \frac{\Delta}{2} \end{pmatrix} \begin{pmatrix} \psi_e(n-1) \\ \psi_g(n) \end{pmatrix} \quad (6.55)$$

This is the same equation as for a classical e.m. field, so that the eigenvalues are easily found to be

$$E_{n,\pm} = (n - \frac{1}{2})\hbar\omega_L \pm \hbar \sqrt{\left(\frac{\Delta}{2}\right)^2 + |\Omega_n|^2} \quad (6.56)$$

The energy also depends on n which means that the phases of states with different n evolve differently ($e^{-iE_{n,\pm}t/\hbar}$). This leads to destructive interference between their amplitudes and to collapse and revival of the excitation probability.

References: [6], [7]

6.4 Models with few levels, dark states, Raman transitions

The most common extension of the two-level model is of course the three-level model. Such a system can be achieved by choosing lasers in a suitable way. An example are transitions from $J = 1$ to $J' = 1$, where the transition from J' to J with $m = m' = 0$ is forbidden by a selection rule. Shining two laser beams with polarization σ_+ and σ_- on the atom therefore depopulates the state $m = 0, J = 1$. After this has happened, one obtains a so-called Λ system consisting of the states $|J = 1, m = \pm 1\rangle$ and $|J' = 1, m' = 0\rangle$.

The Hamilton operator of this system in RWA reads

$$\begin{aligned} H &= \begin{pmatrix} E_e & \hbar\Omega_+ e^{-i\omega_+ t} & \hbar\Omega_- e^{-i\omega_- t} \\ \hbar\Omega_+^* e^{i\omega_+ t} & E_+ & 0 \\ \hbar\Omega_-^* e^{i\omega_- t} & 0 & E_- \end{pmatrix} \\ &= E_e \mathbb{1} + \begin{pmatrix} 0 & \hbar\Omega_+ e^{-i\omega_+ t} & \hbar\Omega_- e^{-i\omega_- t} \\ \hbar\Omega_+^* e^{i\omega_+ t} & E_+ - E_e & 0 \\ \hbar\Omega_-^* e^{i\omega_- t} & 0 & E_- - E_e \end{pmatrix} \end{aligned} \quad (6.57)$$

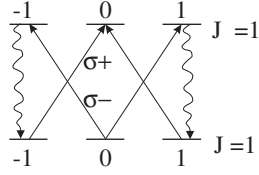


Figure 6.7: The arrows represent the transitions which are driven by the σ_{\pm} light. The eavy lines represent de-excitation due to spontaneous emission.

Again we perform a unitary transformation which removes any time dependence from H :

$$\tilde{\psi}_{\pm} = e^{-i\omega_{\pm}t}\psi_{\pm} \quad (6.58)$$

This leads to the time-independent Hamiltonian

$$H = \hbar \begin{pmatrix} 0 & \Omega_+ & \Omega_- \\ \Omega_+^* & \Delta_+ & 0 \\ \Omega_-^* & 0 & \Delta_- \end{pmatrix} + E_e \mathbb{1} \quad (6.59)$$

where we have set $\Delta_{\pm} = \omega_{\pm} - \frac{E_e - E_{\pm}}{\hbar}$.

6.4.1 Dark States

Consider the case of *two photon resonance*: $\Delta_+ = \Delta_-$. This means that for $\Delta_{\pm} \neq 0$ the individual photons are not in resonance with the respective transitions $|\pm\rangle \rightarrow |e\rangle$. However, we have $\hbar(\omega_+ - \omega_-) = E_- - E_+$ which follows from $\Delta_+ = \Delta_-$ because of wegen

$$\hbar\omega_+ - E_e + E_+ = \hbar\omega_- - E_e + E_-$$

If the condition for two photon resonance is fulfilled, there exists an eigenstate of the Hamiltonian of the form

$$|\psi_D\rangle = \frac{\Omega_-}{\Omega} |+\rangle - \frac{\Omega_+}{\Omega} |-\rangle \quad (6.60)$$

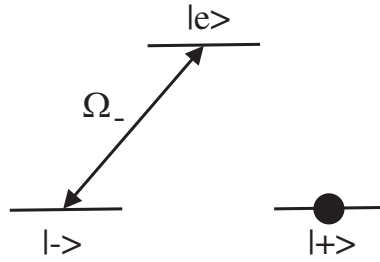
which is called a dark state. Here we have set $\Omega := \sqrt{|\Omega_-|^2 + |\Omega_+|^2}$. The action

of the Hamiltonian on this state is given by

$$\begin{aligned} H |\psi_D\rangle &= \hbar \begin{pmatrix} 0 & \Omega_+ & \Omega_- \\ \Omega_+^* & \Delta_+ & 0 \\ \Omega_-^* & 0 & \Delta_- \end{pmatrix} \begin{pmatrix} 0 \\ \Omega_-/\Omega \\ -\Omega_+/\Omega \end{pmatrix} \\ &= \hbar \begin{pmatrix} \frac{\Omega_+\Omega_-}{\Omega^2} - \frac{\Omega_-\Omega_+}{\Omega^2} \\ \frac{\Delta_+\Omega_-}{\Omega} \\ \frac{-\Delta_-\Omega_+}{\Omega} \end{pmatrix} = \hbar \Delta \begin{pmatrix} 0 \\ \frac{\Omega_-}{\Omega} \\ -\frac{\Omega_+}{\Omega} \end{pmatrix} \end{aligned}$$

for $\Delta_+ = \Delta_- = \Delta$. Hence, in this case $|\psi_D\rangle$ is an eigenstate of H with no excited component. The excitation probability is therefore zero, the atomic gas is for this (!) particular combination of light field transparent.⁵ The transition probabilities interfere destructively.

Simple special case: $\Omega_+ = 0 \Rightarrow |\psi_D\rangle = |+\rangle$

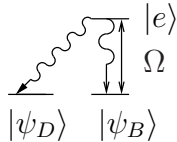


If the Rabi frequencies are changed, $|\psi_D\rangle$ is coupled to the excited state again. For any set of Rabi frequencies, there is a bright state

$$|\psi_B\rangle = \frac{\Omega_+^*}{\Omega} |+\rangle + \frac{\Omega_-^*}{\Omega} |-\rangle \quad (6.61)$$

which is orthogonal to $|\psi_D\rangle$ and is coupled to the excited state, $H |\psi_B\rangle = \Omega |e\rangle + \Delta |\psi_B\rangle$

$|\psi_D\rangle$ and $|\psi_B\rangle$ form an alternative basis for the subspace spanned by $|+\rangle$ and $|-\rangle$. Spontaneous emission from $|e\rangle$ can generally end in $|\psi_D\rangle$ or $|\psi_B\rangle$ since the dark state is only dark for the two laser modes and is coupled to other light modes:



This can be used to pump the atoms into $|\psi_D\rangle$ (see the chapter on laser cooling). For $\Delta_+ \neq \Delta_-$ the dark state $|\psi_D\rangle$ is not an eigenstate of the Hamiltonian

⁵This is also the case when we have no two photon resonance.

anymore and will evolve into the bright state $|\psi_B\rangle$, which can be excited. The pump cycle is then broken.

The physical meaning of dark states can be understood by considering a transition from $J_g = L_g = 1$ to $J_e = L_e = 0$. Although this example is not very realistic (since L_g usually vanishes in the ground state) it displays the main features of dark states.⁶ Consider the form of the electronic states $|e\rangle = |L_e = m_e = 0\rangle$ and $|m\rangle = |L_g = 1, m_g = m\rangle$ in position space. Using spherical coordinates,

$$\mathbf{x} = \begin{pmatrix} r \sin \vartheta \cos \varphi \\ r \sin \vartheta \sin \varphi \\ r \cos \vartheta \end{pmatrix}$$

we find for the states

$$\begin{aligned} \langle \mathbf{x} | -1 \rangle &= Y_{1,-1}(\vartheta, \varphi) r f_g(r) \\ &= r e^{-i\varphi} \sin \vartheta f_g(r) = (x - iy) f_g(r) \\ \langle \mathbf{x} | 0 \rangle &= r \cos \vartheta f_g(r) \\ &= z f_g(r) \\ \langle \mathbf{x} | 1 \rangle &= (x + iy) f_g(r) \\ \langle \mathbf{x} | e \rangle &= Y_{0,0} f_e(r) \\ &= f_e(r) \end{aligned}$$

This results in the following matrix elements of the dipole moment $-e\mathbf{x}$,

$$\begin{aligned} \mathbf{d}_{e,-1} &= \langle e | -e\mathbf{x} | -1 \rangle \\ &= -e \int d^3x f_e^* f_{-1}(r) (x - iy) \begin{pmatrix} x \\ y \\ z \end{pmatrix} \\ &= -e \int d^3x f_e^* f_{-1}(r) \begin{pmatrix} x^2 - ixy \\ xy - iy^2 \\ xz - iyz \end{pmatrix} \end{aligned}$$

By symmetry under $x \rightarrow -x'$ or $y \rightarrow -y'$ one has

$$\mathbf{d}_{e,-1} = -e \int d^3x f_e^*(r) f_{-1} \begin{pmatrix} x^2 \\ -iy^2 \\ 0 \end{pmatrix}$$

⁶In realistic atoms one for instance has $L_g = 0, S = 1, L_e = 1$ so that $J_g = 1$ and $J_e = 0$ for a Λ configuration. Though the angular momenta have just the reversed values as in the example above, selection rules imply that the arguments which will be presented in the following can also be applied to this case.

Symmetry also leads to $\int f(r)x^2 = \int f(r)y^2$, so that $\mathbf{d}_{e,-1} \sim (1, -i, 0)$. Analogously one can show that $\mathbf{d}_{e,1} \sim (1, i, 0)$ and $\mathbf{d}_{e,0} \sim (0, 0, 1)$.

We now focus on the example of light that is linearly polarized along the x direction, so that $\mathbf{E} \sim \mathbf{e}_x$. Because

$$\boldsymbol{\epsilon}_+ + \boldsymbol{\epsilon}_- = \frac{1}{\sqrt{2}}(\mathbf{e}_x + i\mathbf{e}_y) + \frac{1}{\sqrt{2}}(\mathbf{e}_x - i\mathbf{e}_y) = \sqrt{2}\mathbf{e}_x$$

this is equivalent to using left- and right-circularly polarized light of same intensity. This leads to the Rabi frequencies

$$\begin{aligned} \Rightarrow \Omega_+ &= \frac{1}{\hbar} \mathbf{d}_{e,+1} \cdot \mathbf{E} \sim \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix} \cdot \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = 1 \\ \Omega_- &= \frac{1}{\hbar} \mathbf{d}_{e,-1} \cdot \mathbf{E} \sim \begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix} \cdot \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = 1 \\ &\Rightarrow \Omega_+ = \Omega_- \end{aligned}$$

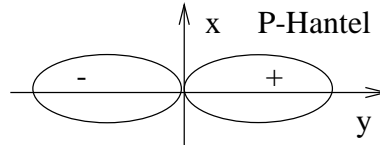
The Rabi frequencies are therefore equal, so that the dark state (6.60) reads

$$|\psi_D\rangle = \frac{1}{\sqrt{2}}(|+1\rangle - |-1\rangle)$$

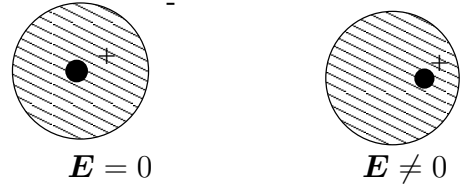
In position space this becomes

$$\begin{aligned} \psi_D(x) &= \frac{1}{\sqrt{2}}(\psi_{+1}(x) - \psi_{-1}(x)) \\ &= \frac{1}{\sqrt{2}}\{(x + iy)f_g(r) - (x - iy)f_g(r)\} \\ &= \sqrt{2}iyf_g(r) \end{aligned} \tag{6.62}$$

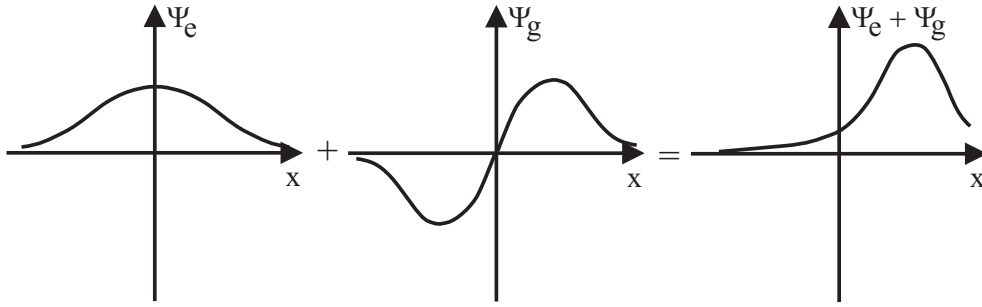
$\Rightarrow \psi_D$ is antisymmetric in y direction and corresponds to a p state along the y axis.



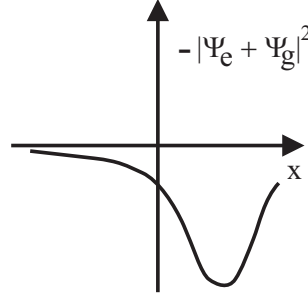
With the knowledge of the spatial form of the dark state we reconsider the interaction between the atoms and the laser beams. Classically, an electric field interacts with an atom by displacing positive and negative charges against each other *along the direction of \mathbf{E}* :



The initially symmetric charge distribution thus becomes asymmetric. In quantum mechanics, one has a similar picture. E creates a superposition of $|e\rangle$ and $|g\rangle$. In our example, $|e\rangle$ is symmetric in position space, and all ground states are antisymmetric. ⁷ A general superposition is not symmetric.



The charge distribution of the electrons is given by $\varrho(\mathbf{x}) = -e|\psi(\mathbf{x})|^2$. It is symmetric both for the excited state $|e\rangle$ and for each of the ground states. Only a superposition corresponds to an asymmetric, displaced charge distribution.



The superposition of symmetric and antisymmetric states is therefore the quantum mechanical description of the displacement of charges in an electric field.

If we apply a laser beam (whose electric field is in the direction of its polarization) to normal atoms, it is possible to find a combination of an s and a p state which corresponds to a charge displacement *along the direction of the electric field*. For atoms in a dark state this is impossible: in our example the electric field is along the x axis, but the dark state (6.62) is antisymmetric along the y -axis. This means that for atoms in a dark state, the charge distribution cannot be displaced along the direction of the electric field.

⁷Just as a reminder: for real atoms the situation is usually inverted.

6.4.2 Raman transitions

Consider the time evolution generated by the Hamiltonian

$$H = \hbar \begin{pmatrix} 0 & \Omega_+ & \Omega_- \\ \Omega_+^* & \Delta_+ & 0 \\ \Omega_-^* & 0 & \Delta_- \end{pmatrix} \quad (6.63)$$

$$\begin{aligned} \Rightarrow i\dot{\psi}_e &= \Omega_+ \psi_+ + \Omega_- \psi_- \\ i\dot{\psi}_{\pm} &= \Delta_{\pm} \psi_{\pm} + \Omega_{\pm}^* \psi_e \end{aligned}$$

Ansatz: $\psi_{\pm} = e^{-i\Delta_{\pm}t} \tilde{\psi}_{\pm} \Rightarrow$

$$i\dot{\psi}_e = \Omega_+ e^{-i\Delta_+t} \tilde{\psi}_+ + \Omega_- e^{-i\Delta_-t} \tilde{\psi}_- \quad (6.64)$$

$$i\dot{\tilde{\psi}}_{\pm} = \Omega_{\pm}^* e^{i\Delta_{\pm}t} \psi_e \quad (6.65)$$

Since $|\psi_e| \leq 1$ one finds that $|\dot{\tilde{\psi}}_{\pm}| \leq |\Omega_{\pm}|$ holds. The formal solution for ψ_e is given by

$$\psi_e(t) = \underbrace{\psi_e(0)}_{\equiv 0} - i \int_0^t dt' \left\{ \Omega_+ e^{-i\Delta_+t'} \tilde{\psi}_+(t') + \Omega_- e^{-i\Delta_-t'} \tilde{\psi}_-(t') \right\} \quad (6.66)$$

We now focus on the case of large detunings $|\Delta_{\pm}| \gg |\Omega_{\pm}|$. In this case, $\tilde{\psi}_{\pm}(t)$ varies slowly compared to $e^{-i\Delta_{\pm}t}$ since $|\dot{\tilde{\psi}}_{\pm}| \leq |\Omega_{\pm}| \ll |\Delta_{\pm}|$. We therefore can set $\tilde{\psi}_{\pm}(t') \approx \tilde{\psi}_{\pm}(t)$ in the integral, which results in

$$\psi_e(t) \approx \frac{\Omega_+}{\Delta_+} (e^{-i\Delta_+t} - 1) \tilde{\psi}_+(t) + \frac{\Omega_-}{\Delta_-} (e^{-i\Delta_-t} - 1) \tilde{\psi}_-(t)$$

Inserting this into Eq. (6.65) leads to

$$i\dot{\tilde{\psi}}_{\pm} = \Omega_{\pm}^* e^{i\Delta_{\pm}t} \left\{ \frac{\Omega_+}{\Delta_+} (e^{-i\Delta_+t} - 1) \tilde{\psi}_+(t) + \frac{\Omega_-}{\Delta_-} (e^{-i\Delta_-t} - 1) \tilde{\psi}_-(t) \right\}$$

or, explicitly for one component,

$$i\dot{\tilde{\psi}}_+ = \frac{|\Omega_+|^2}{\Delta_+} \underbrace{(1 - e^{i\Delta_+t})}_{\approx 1} \tilde{\psi}_+(t) + \frac{\Omega_+^* \Omega_-}{\Delta_-} \underbrace{(e^{i(\Delta_+ - \Delta_-)t} - e^{i\Delta_+t})}_{\approx e^{i(\Delta_+ - \Delta_-)t}} \tilde{\psi}_-(t)$$

where fastly oscillating terms average to zero and therefore can be neglected against the slow terms.⁸ In summary, we thus can write down an effective two-level description for the two ground states,

$$i\hbar \frac{\partial}{\partial t} \begin{pmatrix} \tilde{\psi}_+ \\ \tilde{\psi}_- \end{pmatrix} = H_{\text{eff}} \begin{pmatrix} \tilde{\psi}_+ \\ \tilde{\psi}_- \end{pmatrix} \quad (6.67)$$

with

$$H_{\text{eff}} = \hbar \begin{pmatrix} \frac{|\Omega_+|^2}{\Delta_+} & \frac{\Omega_+^* \Omega_-}{\Delta_-} e^{i(\Delta_+ - \Delta_-)t} \\ \frac{\Omega_-^* \Omega_+}{\Delta_+} e^{-i(\Delta_+ - \Delta_-)t} & \frac{|\Omega_-|^2}{\Delta_-} \end{pmatrix} \quad (6.68)$$

The excited state is never significantly populated and could be omitted in these equations. This procedure is called adiabatic elimination of the excited state.

The matrix elements of H_{eff} are proportional to Ω^2/Δ . It is of fundamental importance that this expression can be large even when the excitation probability $P_e = \Omega^2/\Delta^2$ is small. This makes Raman transitions an excellent tool to coherently manipulate atomic states, since a small excitation probability implies that loss of coherence due to spontaneous emission is strongly suppressed. The time scale of loss of coherence (decoherence) due to spontaneous emission is given by the excitation probability times the rate of spontaneous emission, γP_e . Typical numbers for Raman transitions are $\Omega = 10^9$ Hz, $\gamma = 10^7$ Hz, $\Delta = 10^{10}$ Hz. This results in $\Omega^2/\Delta = 10^8$ Hz and $\gamma P_e = 10^5$ Hz.

The *effective Rabi frequency*

$$\Omega_{\text{eff}} := \frac{\Omega_+^* \Omega_-}{\Delta} \quad (6.69)$$

is a measure for the strength of the coupling between the two ground states. We have omitted the index \pm for the detunings, since for $\Delta_- \neq \Delta_+$ one runs into the problem that H_{eff} is not Hermitean anymore. However, H_{eff} is nevertheless a very useful operator if one knows why this happens. The reason is that the adiabatic evolution of the excited state which was discussed above,

$$\psi_e = \frac{\Omega_+}{\Delta_+} \tilde{\psi}_+ (e^{-i\Delta_+ t} - 1) + \frac{\Omega_-}{\Delta_-} \tilde{\psi}_- (e^{-i\Delta_- t} - 1) \quad (6.70)$$

is actually the first order term of a Taylor expansion of ψ_e with respect to $\frac{1}{\Delta_{\pm}}$. Thus, ψ_e is only correct to first order in $\frac{1}{\Delta_{\pm}}$, if all other frequencies in the Hamiltonian, Ω_{\pm} , $\Delta_+ - \Delta_- \ll \Delta_{\pm}$, are much smaller than the detunings. This implies

⁸In contrast to this, in Eq. (6.65) we had fast terms only. Hence they produce the dominant effect and cannot be neglected.

that we have to do the following expansion:

$$\begin{aligned} \frac{1}{\Delta_-} &= \frac{1}{(\Delta_- - \Delta_+) + \Delta_+} \\ &\approx \frac{1}{\Delta_+} - \frac{1}{\Delta_+^2}(\Delta_- - \Delta_+) + \dots \end{aligned} \quad (6.71)$$

$$= \frac{1}{\Delta_+} + O\left(\frac{1}{\Delta_+^2}\right) \quad (6.72)$$

This means that to first order the expressions $1/\Delta_{\pm}$ are the same, H_{eff} is therefore Hermitean to first order in $\frac{1}{\Delta_{\pm}}$. Introducing the effective Rabi frequency as in (6.69), one can represent H_{eff} as

$$H_{\text{eff}} = \hbar \begin{pmatrix} * & \Omega_{\text{eff}} e^{i(\Delta_+ - \Delta_-)t} \\ \Omega_{\text{eff}}^* e^{-i(\Delta_+ - \Delta_-)t} & * \end{pmatrix} \quad (6.73)$$

This Hamiltonian is analogous to that of a two-level system.

6.5 Adiabatic theorem, STIRAP

The method of **ST**Imulated **R**apid **A**diabatic **P**assage is a highly efficient tool of coherent manipulation which combines dark states and Raman transitions. It is based on the

Adiabatic theorem: Let $H(t)$ be a time-dependent Hamiltonian with instantaneous eigenstates $|n(t)\rangle$, $H(t)|n(t)\rangle = E_n(t)|n(t)\rangle$. Then the evolution of a state $|\psi(t)\rangle$, which initially coincides with the eigenstate $|n(0)\rangle$, (approximately) follows adiabatically $|n(t)\rangle$ if $H(t)$ varies sufficiently slowly, $|\langle n(t)|\dot{n}(t)\rangle| \ll |E_n(t) - E_m(t)|/\hbar$. This means that

$$|\psi(t)\rangle \approx e^{i\phi(t)} |n(t)\rangle \quad (6.74)$$

This is the “folk version” of the theorem. More elaborate versions which also present the correction terms can be found in [4, 40]. The theorem is based on the following observation.

We want to solve $i\hbar\partial_t |\psi(t)\rangle = H(t) |\psi(t)\rangle$ with the initial condition $|\psi(0)\rangle = |n(0)\rangle$. We expand the state in the instantaneous eigenbasis of $H(t)$,

$$|\psi(t)\rangle = \sum_n c_n(t) |n(t)\rangle \quad (6.75)$$

Inserting this into Schrödinger's equation results in

$$\begin{aligned}
 i\hbar\partial_t |\psi\rangle &= i\hbar \sum_n \{\dot{c}_n |n\rangle + c_n |\dot{n}\rangle\} \\
 &= H |\psi\rangle \\
 &= \sum_n E_n(t) c_n(t) |n(t)\rangle
 \end{aligned} \tag{6.76}$$

Multiplying both sides from the left with $\langle m(t)|$, and using $\langle m(t)|n(t)\rangle = \delta_{nm}$ one arrives at

$$i\dot{c}_m + i \sum_n c_n \langle m|\dot{n}\rangle = \frac{E_m}{\hbar} c_m \tag{6.77}$$

Ansatz for the coefficients $c_m(t)$:

$$c_m(t) = \exp\left(-i \int_0^t dt' E_m(t')/\hbar\right) \cdot \tilde{c}_m(t) \tag{6.78}$$

This leads to the following equation for $\dot{\tilde{c}}_m$,

$$\dot{\tilde{c}}_m = - \sum_n \tilde{c}_n \langle m|\dot{n}\rangle \exp\left(-i \int_0^t dt' (E_n(t') - E_m(t'))/\hbar\right) \tag{6.79}$$

If $H(t)$ varies slowly, then $\langle m|\dot{n}\rangle$ is small compared to the phase factor $\int (E_m - E_n)$ for $m \neq n$. We then can neglect these terms and arrive at

$$\dot{\tilde{c}}_m \approx - \langle m|\dot{m}\rangle \tilde{c}_m \tag{6.80}$$

Because

$$\langle m|\dot{m}\rangle = - \langle \dot{m}|m\rangle = -(\langle m|\dot{m}\rangle)^* \tag{6.81}$$

we find that $\langle m|\dot{m}\rangle = i\alpha(t)$ is a purely imaginary number, so that

$$\tilde{c}_m(t) = \exp\left(-i \int_0^t \alpha(t') dt'\right) \tag{6.82}$$

Remark: the phase $\int \alpha(t) dt$ is called *Berry phase* [41] and has a geometric meaning. On a closed path in Hilbert space the Berry phase always has the same value, regardless of whether one traverses the path fast or slow, since Berry's phase is invariant under reparametrizations of time.

6.5.1 Application of the adiabatic theorem to dark states

$$|\psi_D\rangle = \frac{\Omega_-}{\Omega} |+\rangle - \frac{\Omega_+}{\Omega} |-\rangle \quad (6.83)$$

with

$$\Omega = \sqrt{|\Omega_+|^2 + |\Omega_-|^2} \quad (6.84)$$

Let us consider time-dependent Rabi frequencies which are slowly varying, $\Omega_{\pm} = \Omega_{\pm}(t)$. The dark state then also becomes a function of time,

$$|\psi_D\rangle = |\psi_D(T)\rangle . \quad (6.85)$$

We assume that the initial state of the atoms is the initial dark state, $|\psi(0)\rangle = |\psi_D(0)\rangle$. Since the dark state is an eigenstate of the initial Hamiltonian we then find that the state follows adiabatically the evolution of the dark state $|\psi(t)\rangle = |\psi_D(t)\rangle$. This is the principle of STIRAP. With this technique it is possible to transfer atoms from one ground state to another with very high efficiency. More specifically, one does the following.

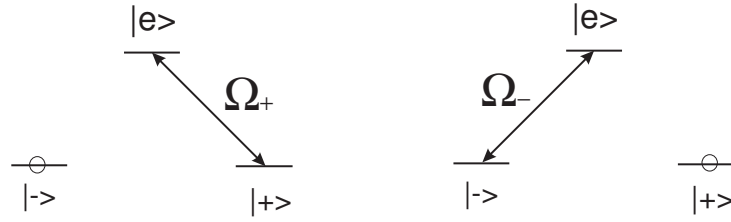


Figure 6.8: The initial state is on the left side, the final state on the right side. The circle indicates the dark state.

- $|\psi(0)\rangle = |-\rangle = |\psi_D\rangle$ with $\Omega_-(0) = 0$
- Change Ω_+ and Ω_- such that $\Omega_+(t_1) = 0$
- Then $|\psi(t_1)\rangle = |+\rangle = |\psi_D(t_1)\rangle$ with $\Omega_+(t_1) = 0$.

This procedure corresponds to a counter-intuitive sequence of two laser pulses where one first applies on that laser pulse which does not couple to the atoms.

In practise, STIRAP has a maximum efficiency of up to 99%. It is remarkable that, from the perspective of the adiabatic theorem, the atoms always remain in a superposition of $|-\rangle$ and $|+\rangle$ ($|\psi_D\rangle$) even though these states are only coupled through $|e\rangle$. In the ideal case (infinitely slow variation) the transfer from $|-\rangle$ to $|+\rangle$ happens without ever exciting the atoms. However, one then also has to wait for an infinitely long time.

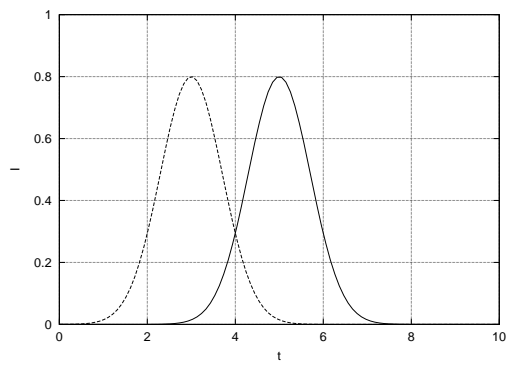


Figure 6.9: The first pulse corresponds to Ω_+ , the second to Ω_- .

Chapter 7

Incoherent Interaction and Density Matrix

7.1 Density Matrix Formalism

The density matrix $\hat{\rho}$ allows us to describe the time evolution of incoherent systems (opposed to pure quantum states). In particular, it also makes it possible to describe open quantum systems.

An example: A (classical) apparatus in a laboratory produces a quantum state, but only with a certain *classical* probability. Each time the experiment is performed, the apparatus may produce a different state with a certain probability. This could for instance be an oven which produces an atom beam sometimes with velocity v_1 and sometimes with v_2 .

One therefore only knows with some probability p_i , $i = 1, 2, \dots$ whether a state $|\psi_i\rangle$ has been prepared. The states $|\psi_i\rangle$ need not to be orthogonal. Of course the classical probabilities must fullfill $\sum_i p_i = 1$, since *some* state has been prepared with certainty. In a closed quantum system (not coupled to a reservoir), each state $|\psi_i\rangle$ then evolves unitarily according to $|\psi_i(t)\rangle = e^{-\frac{i}{\hbar}Ht} |\psi_i(0)\rangle$. If in one run of the experiment the state $|\psi_i\rangle$ has been prepared, the expectation value of any observable for this state is given by

$$\langle \hat{A} \rangle_i = \langle \psi_i(t) | \hat{a} | \psi_i(t) \rangle \quad (7.1)$$

In each run of the experiment, the state $|\psi_i\rangle$ is prepared with probability p_i . One can calculate the full expectation value by averaging over all states which can be prepared,

$$\langle \hat{A} \rangle = \sum_i p_i \langle \hat{A} \rangle_i = \sum_i p_i \langle \psi_i(t) | \hat{a} | \psi_i(t) \rangle \quad (7.2)$$

If $\{|n\rangle\}$ is an orthonormal basis of Hilbert space, one can write the identity operator $\mathbb{1}$ as $\sum_n |n\rangle\langle n|$. We therefore find for the expectation value

$$\langle \hat{A} \rangle = \sum_i p_i \langle \psi_i | \sum_n |n\rangle \langle n| \hat{A} | \psi_i \rangle = \sum_n \langle n | \hat{A} \sum_i p_i | \psi_i \rangle \langle \psi_i | n \rangle = \sum_n \langle n | \hat{A} \hat{\varrho} | n \rangle \quad (7.3)$$

The *Density matrix* $\hat{\varrho}$ is defined by

$$\hat{\varrho} = \sum_i p_i |\psi_i\rangle \langle \psi_i| \quad (7.4)$$

The expectation value of an operator then can be expressed as a trace over the operator and the density matrix,

$$\langle \hat{A} \rangle = \text{Tr} (\hat{\varrho} \hat{A}) \quad (7.5)$$

Properties of the trace

(i) The trace does not depend on the choice of basis.

$$(ii) \text{Tr} (\hat{A} \hat{B}) = \text{Tr} (\hat{B} \hat{A})$$

Proof:

ad (i) Let $\{|n'\rangle\}$ be another basis of Hilbert space. Then

$$\begin{aligned} \text{Tr} (\hat{A}) &= \sum_n \langle n | \hat{A} | n \rangle = \sum_n \langle n | \sum_{n'} |n'\rangle \langle n' | \hat{A} | n \rangle \\ &= \sum_{n'} \langle n' | \hat{A} \underbrace{\sum_n |n\rangle \langle n|}_{\mathbb{1}} | n' \rangle = \sum_{n'} \langle n' | \hat{A} | n' \rangle \end{aligned}$$

ad (ii)

$$\begin{aligned} \text{Tr} (\hat{A} \hat{B}) &= \sum_n \langle n | \hat{A} \hat{B} | n \rangle = \sum_{n, n'} \langle n | \hat{A} | n' \rangle \langle n' | \hat{B} | n \rangle \\ &= \sum_{n, n'} \langle n' | \hat{B} | n \rangle \langle n | \hat{A} | n' \rangle = \sum_{n'} \langle n' | \hat{B} \hat{A} | n' \rangle \end{aligned}$$

Properties of the density matrix

- (i) $\hat{\rho}$ is Hermitean.
- (ii) $\text{Tr } \hat{\rho} = 1$
- (iii) $\text{Tr } (\hat{\rho}^2) \leq 1$ ($= 1$ for pure states)

Proof:

ad (i) trivial because of $p_i \in [0, 1]$

ad (ii)

$$\begin{aligned} \text{Tr } \hat{\rho} &= \sum_{n,i} \langle n | p_i | \psi_i \rangle \langle \psi_i | n \rangle = \sum_i p_i \langle \psi_i | \sum_n | n \rangle \langle n | \psi_i \rangle \\ &= \sum_i p_i \underbrace{\langle \psi_i | \psi_i \rangle}_1 = \sum_i p_i = 1 \end{aligned}$$

ad (iii)

$$\begin{aligned} \text{Tr } (\hat{\rho}^2) &= \sum_n \langle n | \sum_{ij} p_i p_j | \psi_i \rangle \langle \psi_i | p_j | \psi_j \rangle \langle \psi_j | n \rangle = \sum_{i,j} p_i p_j \langle \psi_i | \psi_j \rangle \langle \psi_j | \psi_i \rangle \\ &= \sum_{i,j} p_i p_j \underbrace{|\langle \psi_i | \psi_j \rangle|^2}_{\leq 1} \leq \sum_i p_i \sum_j p_j = 1 \end{aligned}$$

For a *pure state* one has:

$$\hat{\rho} = |\psi\rangle \langle \psi|, \quad \hat{\rho}^2 = \hat{\rho}$$

A *mixture* has at least two different states:

$$\hat{\rho} = p_1 |\psi_1\rangle \langle \psi_1| + p_2 |\psi_2\rangle \langle \psi_2| + \dots, \quad p_i \neq 0$$

The physical difference between a mixture of two states and a superposition becomes very clear by considering the following example: Let $\hat{\rho}_G$ describe a mixture:

$$\hat{\rho}_G = \frac{1}{2}(|e\rangle \langle e| + |g\rangle \langle g|)$$

Half of the atoms are excited, and half are in the ground state. Let us calculate the expectation value of the dipole operator, $\mathbf{d} \propto |e\rangle \langle g| + |g\rangle \langle e|$.

$$\begin{aligned} \langle \mathbf{d} \rangle &\propto \text{Tr } (\hat{\rho}_G (|e\rangle \langle g| + |g\rangle \langle e|)) = \frac{1}{2} \text{Tr } (|g\rangle \langle e| + |e\rangle \langle g|) \\ &= \frac{1}{2} \langle e | \dots | e \rangle + \frac{1}{2} \langle g | \dots | g \rangle = 0 \end{aligned}$$

Let $\hat{\varrho}_S$ describe the following superposition,

$$\begin{aligned} |\psi\rangle &= \frac{1}{\sqrt{2}}(|e\rangle + |g\rangle) \\ \hat{\varrho}_S &= |\psi\rangle\langle\psi| = \frac{1}{2}(|e\rangle + |g\rangle)(\langle g| + \langle e|) \\ &= \frac{1}{2}(\underbrace{|g\rangle\langle g| + |e\rangle\langle e|}_{\hat{\varrho}_G}) + \frac{1}{2}(|e\rangle\langle g| + |g\rangle\langle e|) \end{aligned}$$

$\hat{\varrho}_S$ has non-diagonal matrix elements, which are called *coherences*. The expectation value of the dipole operator is given by

$$\begin{aligned} \langle d \rangle &\propto \text{Tr}(\hat{\varrho}_S(|e\rangle\langle g| + |g\rangle\langle e|)) = \text{Tr}((\hat{\varrho}_G + \frac{1}{2}(|e\rangle\langle g| + |g\rangle\langle e|))(|e\rangle\langle g| + |g\rangle\langle e|)) \\ &= 0 + \frac{1}{2}\text{Tr}(\underbrace{|e\rangle\langle e| + |g\rangle\langle g|}_1) = 1 \end{aligned}$$

The non-diagonal elements of the density matrix describe the degree of quantum mechanical coherence of the system, i.e., its ability to show interference effects. This is the reason why they are called coherences. The diagonal elements are equivalent to the population of each state.

7.2 Liouville Equation, Superoperators

We consider the time evolution of $\hat{\varrho}$ in a closed quantum system. A pure state of course fulfills the Schrödinger equation:

$$i\hbar\partial_t |\psi_i\rangle = H |\psi_i\rangle$$

This can be used to derive an equation for the density matrix:

$$\begin{aligned} i\hbar\partial_t \hat{\varrho} &= i\hbar \sum_i p_i \partial_t (|\psi_i\rangle\langle\psi_i|) = i\hbar \sum_i p_i (\partial_t |\psi_i\rangle \langle\psi_i| + |\psi_i\rangle \partial_t \langle\psi_i|) \\ &= \sum_i p_i (H |\psi_i\rangle\langle\psi_i| - |\psi_i\rangle\langle\psi_i| H) \\ &\quad \boxed{i\hbar\partial_t \hat{\varrho} = [H, \hat{\varrho}]} \end{aligned} \tag{7.6}$$

This is the Liouville equation for closed systems.
The trace of the density matrix is time independent:

$$i\hbar\partial_t \text{Tr} \hat{\varrho} = \text{Tr}([H, \hat{\varrho}]) = \text{Tr}(H\hat{\varrho} - \hat{\varrho}H) = 0$$

$[H, \cdot]$ is a special cases of so-called superoperators, which act on operators instead of states.

$$\mathcal{L}\hat{\varrho} := [H, \hat{\varrho}]$$

is called *Liouville”=Superoperator*. Let us now solve the Liouville equation $i\hbar\partial_t\hat{\varrho} = \mathcal{L}\hat{\varrho}$. Formally this is quickly done:

$$\begin{aligned}\hat{\varrho}(t) &= e^{-\frac{i}{\hbar}t\mathcal{L}}\hat{\varrho}(0) = (1 - \frac{i}{\hbar}t\mathcal{L} - \frac{t^2}{2!\hbar^2}\mathcal{L}\mathcal{L} + \dots)\hat{\varrho}(0) \\ &= \hat{\varrho}(0) - \frac{it}{\hbar}[H, \hat{\varrho}(0)] - \frac{t^2}{2!\hbar^2}[H, [H, \hat{\varrho}(0)]] + \dots = e^{-\frac{i}{\hbar}Ht}\hat{\varrho}(0)e^{\frac{i}{\hbar}Ht}\end{aligned}$$

Proof:

$$f(t) := e^{-\frac{it}{\hbar}\mathcal{L}}\hat{\varrho}(0) \rightarrow i\hbar\partial_t f = \mathcal{L}f = [H, f]$$

$$g(t) := e^{-\frac{i}{\hbar}Ht}\hat{\varrho}(0)e^{\frac{i}{\hbar}Ht}$$

$$\text{to show: } f(t) = g(t)$$

$$i\hbar\partial_t g = i\hbar(-\frac{i}{\hbar}Hg + \frac{i}{\hbar}gH) = [H, g]$$

$f(t)$ and $g(t)$ fullfill the same differential equation.

$$f(0) = \hat{\varrho}(0) = g(0)$$

$f(t)$ and $g(t)$ fullfill the same initial conditions.

$$\rightarrow f(t) = g(t)$$

Entropy, Equilibrium Density Matrix (This is a short deviation to statistical mechanics)

The entropy of a quantum system is defined by

$$S = -k_B \text{Tr}(\hat{\varrho} \ln \hat{\varrho}) = -k_B \langle \ln \hat{\varrho} \rangle \quad (7.7)$$

S describes the degree of order in a system.

In (7.7) we take the logarithm of the density matrix. Generally, a function $f(\hat{A})$ of an operator is defined by refering to its diagonal representation.

$$\hat{A}|a_n\rangle = a_n|a_n\rangle \rightarrow \hat{A} = \sum_n a_n|a_n\rangle\langle a_n|$$

For Hermitean operators we have $\langle a_n|a_m\rangle = \delta_{nm}$

f should be expandable into a Taylor series,

$$\begin{aligned}f(\hat{A}) &= \sum_l \frac{f_l}{l!} \hat{A}^l \\ \hat{A}^2 &= \sum_n a_n|a_n\rangle\langle a_n| \sum_m a_m|a_m\rangle\langle a_m| = \sum_n a_n^2|a_n\rangle\langle a_n|\end{aligned}$$

This can be generalized to arbitrary powers:

$$\begin{aligned}\hat{A}^l &= \sum_n a_n^l |a_n\rangle\langle a_n| \\ \Rightarrow f(\hat{A}) &= \sum_l \frac{f_l}{l!} \sum_n a_n^l |a_n\rangle\langle a_n| = \sum_n f(a_n) |a_n\rangle\langle a_n|\end{aligned}\quad (7.8)$$

We can employ this to calculate the entropy of a pure state:

$$\begin{aligned}\hat{\varrho} &= 1 \cdot |\psi\rangle\langle\psi| \\ S &= -k_B \text{Tr}(\hat{\varrho} \ln \hat{\varrho}) = -k_B \text{Tr}(1 \cdot |\psi\rangle\langle\psi| \cdot \underbrace{\ln(1)}_{=0} |\psi\rangle\langle\psi|)\end{aligned}$$

The entropy of a pure state is 0.

For a system in equilibrium we have

$$\dot{\hat{\varrho}} = 0 \Leftrightarrow [\hat{H}, \hat{\varrho}] = 0$$

$\Rightarrow \hat{\varrho}$ has the same eigenvectors as \hat{H}

$$\begin{aligned}H |E_n\rangle &= E_n |E_n\rangle \\ \Rightarrow \hat{\varrho} &= \sum_n \varrho_n |E_n\rangle\langle E_n|\end{aligned}$$

Thermal equilibrium: maximise the entropy S under the constraints

- $\text{Tr}(\hat{\varrho}) = 1$ (Conservation of total probability)
- $\langle \hat{H} \rangle = E$ (For canonical or grand canonical ensemble)

To do so we introduce two Lagrange parameters λ_1, λ_2 . Maximising results in

$$\frac{\partial}{\partial \varrho_n} \left(S - \lambda_1 (\text{Tr}(\hat{\varrho}) - 1) - \lambda_2 (\langle \hat{H} \rangle - E) \right) = 0$$

Since $\hat{\varrho}$ is diagonal in this basis we find

$$\begin{aligned}\frac{\partial}{\partial \varrho_n} \left\{ -k_B \sum_{n'} \varrho_{n'} \ln \varrho_{n'} - \lambda_1 \left(\sum_{n'} \varrho_{n'} - 1 \right) - \lambda_2 \left(\sum_{n'} \varrho_{n'} E_{n'} - E \right) \right\} &= 0 \\ \Rightarrow 0 &= -k_B (\ln \varrho_n + 1) - \lambda_1 - \lambda_2 E_n\end{aligned}$$

$$\begin{aligned}\Rightarrow \varrho_n &= \exp \left\{ -\frac{1}{k_B} (\lambda_1 + \lambda_2 E_n) \right\} \\ &\sim e^{-\frac{\lambda_2}{k_B} E_n}\end{aligned}$$

λ_1 is fixed by $\sum_n \varrho_n = 1$

$$\Rightarrow \hat{\varrho} \sim \sum_n e^{-\frac{\lambda_2}{k_B} E_n} |E_n\rangle \langle E_n|$$

Consider this as a function of H

$$\hat{\varrho} \sim e^{-\frac{\lambda_2}{k_B} H}$$

$$\text{Let } \lambda_2 = \frac{1}{T} \Rightarrow \hat{\varrho} = \frac{1}{Z} e^{-\beta \hat{H}}, \beta = \frac{1}{k_B T}, Z = \text{Tr} \left(e^{-\beta \hat{H}} \right)$$

Liouville's Theorem The entropy of a system is not changed if a unitary transformation is applied to the density matrix operator.

$$\hat{\varrho}' = U \hat{\varrho} U^\dagger$$

$$S' = -k_B \text{Tr} (\hat{\varrho}' \ln \hat{\varrho}') = S(\hat{\varrho}') = S(U \hat{\varrho} U^\dagger)$$

Taylor expansion of the operator inside the trace:

$$\hat{K}(\hat{\varrho}') := \hat{\varrho}' \ln \hat{\varrho}' = \sum_n K_n \hat{\varrho}'^n = \sum_n K_n (U \hat{\varrho} U^\dagger)^n$$

Evaluate the term in parentheses,

$$\begin{aligned}(U \hat{\varrho} U^\dagger)^n &= U \hat{\varrho} \underbrace{U^\dagger U}_1 \hat{\varrho} U^\dagger U \hat{\varrho} U^\dagger \dots \\ &= U \hat{\varrho}^n U^\dagger\end{aligned}$$

$$\Rightarrow \hat{K}(\hat{\varrho}') = U \sum K_n \hat{\varrho}^n U^\dagger$$

$$\begin{aligned}\Rightarrow S' &= -k_B \text{Tr} (U \hat{\varrho} \ln \hat{\varrho} U^\dagger) \\ &= -k_B \text{Tr} (U^\dagger U \hat{\varrho} \ln \hat{\varrho}) = S\end{aligned}$$

\Rightarrow The entropy cannot be changed by the time evolution $U = \exp(-iHt/\hbar)$ in a closed system. If one wants to change it, the quantum system must be open. This means that the system can exchange energy, momentum, or particles by a coupling to the environment or reservoir. This is exploited in cooling of quantum systems.

7.3 Reduced density matrix, Zwanzig's Master Equation

Direct product of Hilbert spaces Consider two Hilbert spaces \mathcal{H}_S , \mathcal{H}_R with bases $\{|S_n\rangle\}$, $\{|R_\alpha\rangle\}$. Their direct product is given by

$$\mathcal{H}_S \otimes \mathcal{H}_R = \left\{ |\psi\rangle \left| \begin{array}{l} |\psi\rangle = \sum_{n,\alpha} \psi_{n\alpha} |S_n\rangle \otimes |R_\alpha\rangle \end{array} \right. \right\}$$

This space does not only contain product states of the form $|S_n\rangle \otimes |R_\alpha\rangle$, but also *entangled states*, z.B.

$$|S_1\rangle \otimes |R_1\rangle + |S_2\rangle \otimes |R_2\rangle$$

Entangled states play a very important role in many modern areas of Physics (Quantum information, Bell inequalities, Quantum computing, ..). They are special with respect to the correlations between the two subsystems R and S . Correlations in entangled states cannot be explained by any classical model. More on this topic can be found in Ref. [19].

Let $\hat{\rho}$ be the density matrix in the space $\mathcal{H}_S \otimes \mathcal{H}_R$. The expectation value of operators which only act on states in \mathcal{H}_S is given by $\langle \hat{A}_S \rangle = \text{Tr} \left(\hat{\rho} \left(\hat{A}_S \otimes \mathbf{1}_R \right) \right)$.

Here, the operator \hat{A}_S acts only on \mathcal{H}_S :

$$\hat{A}_S : \mathcal{H}_S \rightarrow \mathcal{H}_S : |S_n\rangle \mapsto \hat{A}_S |S_n\rangle$$

$\hat{A}_S \otimes \mathbf{1}_R$ is the representation of this operator on the product Hilbert space. It acts like

$$\begin{aligned} \hat{A}_S \otimes \mathbf{1}_R : \mathcal{H}_S \otimes \mathcal{H}_R &\rightarrow \mathcal{H}_S \otimes \mathcal{H}_R \\ |S_n\rangle \otimes |R_\alpha\rangle &\mapsto \hat{A}_S |S_n\rangle \otimes \mathbf{1}_R |R_\alpha\rangle \end{aligned}$$

7.3.1 Reduced Density Matrix, Projection Superoperators

The trace of an operator in product space can therefore be decomposed as

$$\text{Tr}(\hat{O})_{\mathcal{H}_S \otimes \mathcal{H}_R} = \sum_{n,\alpha} (\langle S_n| \otimes \langle R_\alpha|) \hat{O} (|S_n\rangle \otimes |R_\alpha\rangle)$$

\Rightarrow

$$\begin{aligned} \langle \hat{A}_S \rangle &= \sum_{n,\alpha} \langle S_n| \otimes \langle R_\alpha| \hat{\rho} (\hat{A}_S \otimes \mathbf{1}_R) |S_n\rangle \otimes |R_\alpha\rangle \\ &= \sum_n \langle S_n| \sum_\alpha \langle R_\alpha| \hat{\rho} |R_\alpha\rangle \hat{A}_S |S_n\rangle \\ &= \sum_n \langle S_n| \text{Tr}_R(\hat{\rho}) \hat{A}_S |S_n\rangle \end{aligned}$$

In the last step we have used that the operator acts only on one subspace.

$$\text{Tr}_R(\hat{\varrho}) \equiv \sum_{\alpha} \langle R_{\alpha} | \hat{\varrho} | R_{\alpha} \rangle$$

Define

$$\boxed{\hat{\varrho}_S = \text{Tr}_R(\hat{\varrho})}$$

reduced density matrix.

$\hat{\varrho}_S$ act on \mathcal{H}_S . In particular we have

$$\langle \hat{A}_S \rangle = \text{Tr}_S(\hat{\varrho}_S \hat{A}_S) = \text{Tr}_S(\text{Tr}_R(\hat{\varrho}) \hat{A}_S)$$

Obviously, the reduced density matrix contains the full information about the expectation values of all observables on the system Hilbert space \mathcal{H}_S .

Aim: Find an equation for the evolution of $\hat{\varrho}_S$ alone.

Projection Superoperators In quantum mechanics, ordinary projection operators \hat{P} , like $P = |e\rangle\langle e|$ with $P|\psi\rangle = |e\rangle\langle e|\psi\rangle$, fulfill $P^2 = P$, $P^\dagger = P$

Analogously we find for the action of a projection superoperator on an operator

$$\mathcal{P}^2 \hat{A} = \mathcal{P} \hat{A}$$

Examples:

1. Reduced density matrix

$$\mathcal{P} \hat{\varrho} \equiv \hat{\varrho}_R(0) \text{Tr}_R(\hat{\varrho}), \quad (7.9)$$

where $\hat{\varrho}_R(0)$ corresponds to the density matrix of the reservoir at time $t = 0$

$$\begin{aligned} \mathcal{P}^2 \hat{\varrho} &= \hat{\varrho}_R(0) \text{Tr}_R(\mathcal{P} \hat{\varrho}) \\ &= \hat{\varrho}_R(0) \text{Tr}_R(\hat{\varrho}_R(0) \underbrace{\text{Tr}_R(\hat{\varrho})}_{\hat{\varrho}_S: \mathcal{H}_S \mapsto \mathcal{H}_S}) \end{aligned}$$

\Rightarrow

$$\mathcal{P}^2 \hat{\varrho} = \hat{\varrho}_R(0) \text{Tr}_R(\hat{\varrho}_R(0)) \hat{\varrho}_S = \mathcal{P} \hat{\varrho}$$

2. Projection on the populations

$$\mathcal{P} \hat{\varrho} \equiv \sum_n |n\rangle \langle n| \hat{\varrho} |n\rangle \langle n|$$

\mathcal{P} projects $\hat{\varrho}$ on its diagonal elements, i.e., all coherences are removed.

7.3.2 Zwanzig's Master Equation

Zwanzig's Master equation is an equation for $\mathcal{P}\hat{\rho}$ alone, which is derived from Liouville's equation

$$i\hbar\dot{\rho} = \mathcal{L}\rho$$

for $\hat{\rho}$. To do so we first introduce a second projection superoperator $\mathcal{Q} := 1 - \mathcal{P}$, which fullfills $\mathcal{Q}^2 = \mathcal{Q}$.¹ The projection superoperator is time-independent, $\dot{\mathcal{P}} = 0$,

$$\begin{aligned} \Rightarrow i\hbar\frac{\partial}{\partial t}\mathcal{P}\rho &= \mathcal{P}\mathcal{L}\rho = \mathcal{P}\mathcal{L}(\mathcal{P} + \mathcal{Q})\rho \\ \Rightarrow i\hbar\frac{\partial}{\partial t}\mathcal{P}\rho &= \mathcal{P}\mathcal{L}\mathcal{P}\rho + \mathcal{P}\mathcal{L}\mathcal{Q}\rho \end{aligned}$$

analogously with $\mathcal{Q}\rho$

$$i\hbar\frac{\partial}{\partial t}\mathcal{Q}\rho = \mathcal{Q}\mathcal{L}\mathcal{P}\rho + \mathcal{Q}\mathcal{L}\mathcal{Q}\rho \quad (7.10)$$

First we solve $i\hbar\mathcal{Q}\dot{\rho} = \mathcal{Q}\mathcal{L}\mathcal{Q}\rho$. When the superoperators are time-independent, the formal solution is easily given:

$$\mathcal{Q}\rho(t) = \exp(-i\mathcal{Q}\mathcal{L}\mathcal{Q}t/\hbar)\mathcal{Q}\rho(0) .$$

The general case is considerably more complicated and leads to a so-called time-ordered exponential. Since this appears *very* often in the interaction picture and in quantum field theory, we will consider this in more detail here.

The Time-Ordered Exponential

Formal solution:

$$i\hbar \int_0^t \mathcal{Q}\dot{\rho} dt = \int_0^t \mathcal{Q}\mathcal{L}(t')\mathcal{Q}\rho(t') dt' \quad (7.11)$$

$$\Rightarrow \mathcal{Q}\rho(t) = \mathcal{Q}\rho(0) - \frac{i}{\hbar} \int_0^t \mathcal{Q}\mathcal{L}(t')\mathcal{Q}\rho(t') dt' \quad (7.12)$$

where in the last step we used $\mathcal{Q}^2 = \mathcal{Q}$. This solution is only formal since it contains itself: $\mathcal{Q}\rho(t)$ depends on $\mathcal{Q}\rho(t')$ with $t' < t$. If $\mathcal{Q}\rho(t')$ is rewritten using again Eq. (7.12), one arrives at

$$\mathcal{Q}\rho(t) = \mathcal{Q}\rho(0) - \frac{i}{\hbar} \int_0^t dt' (\mathcal{Q}\mathcal{L}(t')\mathcal{Q}) \left\{ \mathcal{Q}\rho(0) - \frac{i}{\hbar} \int_0^{t'} dt'' \mathcal{Q}\mathcal{L}(t'')\mathcal{Q}\rho(t'') \right\} \quad (7.13)$$

¹There is an important difference between projection operators and projection superoperators. For projection operators one has $|\psi\rangle = \begin{pmatrix} P|\psi\rangle \\ Q|\psi\rangle \end{pmatrix}$. However, for superoperators we have $\hat{A} =$

$$\begin{pmatrix} \mathcal{P}\hat{A}\mathcal{P} & \mathcal{P}\hat{A}\mathcal{Q} \\ \mathcal{Q}\hat{A}\mathcal{P} & \mathcal{Q}\hat{A}\mathcal{Q} \end{pmatrix}$$

Iteration leads to

$$\begin{aligned} \Rightarrow \mathcal{Q}_\rho(t) &= \mathcal{Q}_\rho(0) - \frac{i}{\hbar} \int_0^t dt' (\mathcal{Q}\mathcal{L}(t') \mathcal{Q}) \mathcal{Q}_\rho(0) \\ &+ \left(-\frac{i}{\hbar}\right)^2 \int_0^t dt' \int_0^{t'} dt'' (\mathcal{Q}\mathcal{L}\mathcal{Q}(t')) (\mathcal{Q}\mathcal{L}\mathcal{Q}(t'')) \mathcal{Q}_\rho(0) + \dots \end{aligned} \quad (7.14)$$

We introduce the *time-ordered product* :

$$T(A(t)B(t')) := \begin{cases} A(t)B(t') & \text{for } t > t' \\ B(t')A(t) & \text{for } t' > t \end{cases} \quad (7.15)$$

This allows us to rewrite the multiple integrals in the series in the following way:

$$\int_0^t dt_1 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \int_0^{t_1} dt_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) = \int_0^t dt_2 \int_{t_2}^t dt_1 T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2)) \quad (7.16)$$

$T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2))$ has also the correct ordering of the operators (the operator at a later time is to the left of that at an earlier time) if $t_1 < t_2$. Consider therefore the following integral with modified boundary values for t_2 :

$$\begin{aligned} \Rightarrow & \int_0^t dt_1 \int_0^t dt_2 T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2)) \\ &= \int_0^t dt_1 \int_0^{t_1} dt_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) + \int_0^t dt_1 \int_{t_1}^t dt_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \end{aligned} \quad (7.17)$$

The first term on the right-hand side (rhs) of Eq. (7.17) appears in the series (Eq. (7.14)) vor. The second term on the rhs of Seite von Eq. (7.17) can be written as

$$\int_0^t dt_1 \int_{t_1}^t dt_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) = \int_0^t dt_2 \int_0^{t_2} dt_1 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) . \quad (7.18)$$

With $t'_1 = t_2$ und $t'_2 = t_1$ it follows that

$$\Rightarrow \text{2ndTerm in Eq. (7.17)} = \int_0^t dt'_1 \int_0^{t'_1} dt'_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t'_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t'_2) \quad (7.19)$$

Eq. (7.19) is again a term which appears in the series Reihe (Eq. (7.14)).

$$\begin{aligned} \Rightarrow & \int_0^t dt_1 \int_0^{t_1} dt_2 \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2) \\ &= \frac{1}{2!} \int_0^t dt_1 \int_0^t dt_2 T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \mathcal{Q}\mathcal{L}\mathcal{Q}(t_2)) \end{aligned} \quad (7.20)$$

We thus have found an expression for the first term in the series in which the integrals have independent boundaries. It can be shown that generally

$$\begin{aligned} & \int_0^t dt_1 \int_0^{t_1} dt_2 \dots \int_0^{t_{n-1}} dt_n \mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \dots \mathcal{Q}\mathcal{L}\mathcal{Q}(t_n) \\ &= \frac{1}{n!} \int_0^t dt_1 \dots \int_0^t dt_n T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \dots \mathcal{Q}\mathcal{L}\mathcal{Q}(t_n)) \end{aligned} \quad (7.21)$$

$$\begin{aligned} \Rightarrow \mathcal{Q}_\varrho(t) &= \left\{ 1 - \frac{i}{\hbar} \int_0^t dt_1 \mathcal{Q}\mathcal{L}\mathcal{Q} \dots \right. \\ &\quad \left. + \frac{1}{n!} \left(\frac{-i}{\hbar} \right)^n \int_0^t dt_1 \dots \int_0^t dt_n T(\mathcal{Q}\mathcal{L}\mathcal{Q}(t_1) \dots \mathcal{Q}\mathcal{L}\mathcal{Q}(t_n)) \right\} \mathcal{Q}_\varrho(0) \\ &= T \exp \left(\frac{-i}{\hbar} \int_0^t dt' \mathcal{Q}\mathcal{L}\mathcal{Q}(t') \right) \mathcal{Q}_\varrho(0) \end{aligned} \quad (7.22)$$

$$= : U_{QQ}(t) \mathcal{Q}_\varrho(0) \quad (7.23)$$

This is the solution of the homogeneous differential equation

$$i\hbar \mathcal{Q}\dot{\varrho} = \mathcal{Q}\mathcal{L}\mathcal{Q}(\mathcal{Q}_\varrho) \quad (7.24)$$

The time-ordered exponential $T \exp$ is defined by its Taylor series, in which integrals over different variables t_1, t_2, \dots do appear. The time-ordering operator T guarantees that the operators appear in the same order as in the series (7.14) erscheinen.

Derivation of Master equation continued

To derive the Master equation it is now useful to make the ansatz $\mathcal{Q}_\varrho(t) = U_{QQ}(t) \tilde{\varrho}_Q(t)$.

$$\Rightarrow i\hbar \partial_t \mathcal{Q}_\varrho = i\hbar \dot{U}_{QQ} \tilde{\varrho}_Q + i\hbar U_{QQ} \dot{\tilde{\varrho}}_Q \quad (7.25)$$

With $\dot{U}_{QQ} = -\frac{i}{\hbar} \mathcal{Q}\mathcal{L}\mathcal{Q}(t) U_{QQ}(t)$ and using Eq. (7.10) we find

$$i\hbar \dot{\tilde{\varrho}}_Q = U_{QQ}^{-1}(t) \mathcal{Q}\mathcal{L}\mathcal{P}_\varrho(t) \quad (7.26)$$

$$\Rightarrow \tilde{\varrho}_Q(t) = \tilde{\varrho}_Q(0) - \frac{i}{\hbar} \int_0^t dt' U_{QQ}^{-1}(t') \mathcal{Q}\mathcal{L}\mathcal{P}_\varrho(t') \quad (7.27)$$

$$\Rightarrow \mathcal{Q}_\varrho(t) = \mathcal{Q}_\varrho(0) - \frac{i}{\hbar} U_{QQ}(t) \int_0^t dt' U_{QQ}^{-1}(t') \mathcal{Q}\mathcal{L}\mathcal{P}_\varrho(t') \quad (7.28)$$

Insert Eq. (7.28) into the equation for \mathcal{P}_ϱ :

$$\boxed{i\hbar \mathcal{P}\dot{\varrho} = \mathcal{P}\mathcal{L}\mathcal{P}_\varrho + \mathcal{P}\mathcal{L}(t) \mathcal{Q} \left\{ \mathcal{Q}_\varrho(0) - \frac{i}{\hbar} U_{QQ}(t) \int_0^t dt' U_{QQ}^{-1}(t') \mathcal{Q}\mathcal{L}\mathcal{P}_\varrho(t') \right\}} \quad (7.29)$$

Eq. (7.29) is *Zwanzig's Master equation*.

This is an exact differential equation for $\mathcal{P}\varrho$ alone, but it generally cannot be solved without approximations. To simplify it we now consider the special case of a projection superoperator of the form 7.9 with the following features:

1. $\varrho(0) = \varrho_R(0) \otimes \varrho_S(0)$, i.e., there are no correlation between the system and the reservoir at time $t = 0$
2. $[\varrho_R(0), H_R] = 0$, i.e., the reservoir is in a stationary state of H_R (we set $H = H_R + H_S + H_{\text{int}}$).
3. $\text{Tr}_R(\varrho_R(0)H_{\text{int}}) = 0$ e.g. $H_{\text{int}} = -\mathbf{d} \cdot \mathbf{E}$, here the reservoir corresponds to the radiation field
 $\Rightarrow \text{Tr}_R(\varrho_R(0)H_{\text{int}}) = -\mathbf{d} \langle \mathbf{E} \rangle (t = 0) = 0$.²

When these conditions are valid one finds

$$\begin{aligned}
 \mathcal{Q}\varrho(0) &= \varrho(0) - \mathcal{P}\varrho(0) \\
 &= \varrho_R(0) \otimes \varrho_S(0) - \varrho_R(0) \otimes \text{Tr}_R(\varrho_R(0) \otimes \varrho_S(0)) \\
 &= \varrho_R(0) \otimes \varrho_S(0) - \varrho_R(0) \otimes \varrho_S(0) \\
 &= 0
 \end{aligned} \tag{7.30}$$

The following relations for an arbitrary operator X are left to the reader as an exercise:

$$\mathcal{P}\mathcal{L}\mathcal{P}X = \varrho_R(0)[H_S, \text{Tr}_R(X)] = \mathcal{P}\mathcal{L}_S X \tag{7.31}$$

$$\mathcal{P}\mathcal{L}\mathcal{Q}X = \varrho_R(0)\text{Tr}_R([H_{\text{int}}, X]) = \mathcal{P}\mathcal{L}_{\text{int}}X \tag{7.32}$$

$$\mathcal{Q}\mathcal{L}\mathcal{P}X = [H_{\text{int}}, \varrho_R(0)\text{Tr}_R(X)] = \mathcal{L}_{\text{int}}\mathcal{P}X \tag{7.33}$$

Furthermore,

$$\begin{aligned}
 \mathcal{Q}\mathcal{L}\mathcal{Q}X &= (1 - \mathcal{P})\mathcal{L}\mathcal{Q}X \\
 &= \mathcal{L}\mathcal{Q}X - \mathcal{P}\mathcal{L}\mathcal{Q}X \\
 &= \mathcal{L}(1 - \mathcal{P})X - \mathcal{P}\mathcal{L}_{\text{int}}X \\
 &= \mathcal{L}X - \mathcal{L}\mathcal{P}X - \mathcal{P}\mathcal{L}_{\text{int}}X \\
 &= [H_S + H_R + H_{\text{int}}, X] - [H_S + H_R + H_{\text{int}}, \varrho_R(0)\text{Tr}_R(X)] \\
 &\quad - \varrho_R(0)\text{Tr}_R([H_{\text{int}}, X]) \\
 &= [H_S + H_R + H_{\text{int}}, X] - [H_S, \varrho_R(0)\text{Tr}_R(X)] \\
 &\quad - [H_R, \varrho_R(0)]\text{Tr}_R(X) - [H_{\text{int}}, \varrho_R(0)\text{Tr}_R(X)] \\
 &\quad - \varrho_R(0)\text{Tr}_R([H_{\text{int}}, X])
 \end{aligned}$$

²In a stationary state in free space one has $\langle \mathbf{E} \rangle = 0$, since such a state corresponds to a mixture (i.e., not a superposition) of number states. Because $\mathbf{E} \sim a + a^\dagger$ one then finds $\langle n | a + a^\dagger | n \rangle = \langle n | (\sqrt{n} | n-1 \rangle + \sqrt{n+1} | n+1 \rangle) = 0$

Using $[H_R, \varrho_R(0)] = 0$ one deduces

$$\begin{aligned} \mathcal{Q}\mathcal{L}\mathcal{Q}X &= [H_S, X - \mathcal{P}X] + [H_R, X] + [H_{\text{int}}, X - \mathcal{P}X] - \mathcal{P}[H_{\text{int}}, X] \\ &= [H_S, \mathcal{Q}X] + \mathcal{L}_R X + [H_{\text{int}}, \mathcal{Q}X] - \mathcal{P}\mathcal{L}_{\text{int}}X \\ &= (\mathcal{L}_S \mathcal{Q} + \mathcal{L}_R + \mathcal{L}_{\text{int}} \mathcal{Q} - \mathcal{P}\mathcal{L}_{\text{int}})X \end{aligned} \quad (7.34)$$

Additional properties:

$$\mathcal{L}_S \mathcal{Q} = \mathcal{Q} \mathcal{L}_S \quad (7.35)$$

$$\mathcal{P} \mathcal{L}_S = \mathcal{L}_S \mathcal{P} \quad (7.36)$$

Insertion into the master equation (Eq. (7.29)) results in

$$\begin{aligned} i\hbar \dot{\mathcal{P}} &= \mathcal{P} \mathcal{L}_S \mathcal{P} \varrho - \frac{i}{\hbar} \mathcal{P} \mathcal{L}_{\text{int}} \int_0^t dt' U_{\mathcal{Q}\mathcal{Q}}(t) U_{\mathcal{Q}\mathcal{Q}}^{-1}(t') \mathcal{L}_{\text{int}} \mathcal{P} \varrho(t') \\ U_{\mathcal{Q}\mathcal{Q}}(t) &= T \exp \left(\frac{-i}{\hbar} \int_0^t dt'' \mathcal{Q} \mathcal{L}(t'') \mathcal{Q} \right) \\ \mathcal{L}(t)X &= [H(t), X] = [H_S + H_R + H_{\text{int}}, X] \end{aligned}$$

Approximation: take only terms up to 2nd order in H_{int} into account (assumption: $H_{\text{int}} \ll H_R, H_S$)

$$\Rightarrow U_{\mathcal{Q}\mathcal{Q}} \approx \exp \left(-\frac{i}{\hbar} \mathcal{Q}(\mathcal{L}_S + \mathcal{L}_R) \mathcal{Q} t \right)$$

(if H_S and H_R are time independent).

$$\begin{aligned} \mathcal{Q} \mathcal{L}_S &= \mathcal{L}_S \mathcal{Q} & \Rightarrow \mathcal{Q} \mathcal{L}_S \mathcal{Q} &= \mathcal{L}_S \mathcal{Q} = \mathcal{Q} \mathcal{L}_S \\ \mathcal{Q} \mathcal{L}_R &= \mathcal{L}_R \mathcal{Q} = \mathcal{L}_R \\ &\Rightarrow U_{\mathcal{Q}\mathcal{Q}} = e^{-i\mathcal{Q}\mathcal{L}_S t/\hbar} e^{-i\mathcal{L}_R t/\hbar} \end{aligned}$$

In the last step $\mathcal{L}_S \mathcal{L}_R = \mathcal{L}_R \mathcal{L}_S$ was used, which can be proven in the following way:

$$\begin{aligned} (\mathcal{L}_R \mathcal{L}_S - \mathcal{L}_S \mathcal{L}_R)X &= [H_R, [H_S, X]] - [H_S, [H_R, X]] \\ &= [H_R, [H_S, X]] + [H_S, [X, H_R]] \\ &= -[X, \underbrace{[H_R, H_S]}_{=0}] = 0 \end{aligned}$$

where Jakobi's identity was employed. One then finds

$$e^{-i\mathcal{Q}\mathcal{L}_S t/\hbar} = \sum_{n=0}^{\infty} \frac{(-it/\hbar)^n}{n!} (\mathcal{Q}\mathcal{L}_S)^n \quad (\mathcal{Q}\mathcal{L}_S)^n = \mathcal{L}_S^n \mathcal{Q} = \mathcal{L}_S^n \mathcal{Q} \quad n > 0$$

$$\Rightarrow e^{-i\mathcal{Q}\mathcal{L}_s t/\hbar} \mathcal{Q}\mathcal{L}\mathcal{P} = e^{-i\mathcal{L}_s t/\hbar} \mathcal{Q}\mathcal{L}\mathcal{P}$$

so that the master equation can be cast into the form

$$i\hbar\mathcal{P}\dot{\varrho} = \mathcal{P}\mathcal{L}_S\mathcal{P}\varrho - \frac{i}{\hbar}\mathcal{P}\mathcal{L}_{int} \underbrace{\int_0^t dt' \exp\left\{\frac{-i}{\hbar}(t-t')(\mathcal{L}_s + \mathcal{L}_R)\right\} \mathcal{L}_{int}\mathcal{P}\varrho(t')}_{\text{free evolution}}$$

$$\mathcal{P}\varrho = \varrho_R(0)\text{Tr}_R(\varrho) = \varrho_R(0)\varrho_s(t)$$

$$\begin{aligned} \Rightarrow i\hbar\varrho_R(0)\dot{\varrho}_s &= \varrho_R(0) \underbrace{\text{Tr}_R([H_S, \varrho_R(0)\varrho_s])}_{[H_S, \varrho_s]} - \\ &\quad - \frac{i}{\hbar}\varrho_R(0)\text{Tr}_R\left([H_{int}, \int_0^t dt' e^{-i(t-t')(\mathcal{L}_s + \mathcal{L}_R)/\hbar} [H_{int}, \varrho_R(0)\varrho_s(t')]]\right) \end{aligned}$$

$$i\hbar\dot{\varrho}_s(t) = [H_S, \varrho_s(t)] - \frac{i}{\hbar} \int_0^t dt' \text{Tr}_R\left([H_{int}, e^{-i(t-t')(\mathcal{L}_s + \mathcal{L}_R)/\hbar} [H_{int}, \varrho_R(0)\varrho_s(t')]]\right)$$

This is the general master equation for a system fulfilling $H_{int} \ll H_R, H_S$ and $\dot{H}_s = \dot{H}_R = 0$.

Assumption about the form of the interaction: $H_{int} = -d_i E_i$, $i = 1, \dots, N$ where operators d_i act on \mathcal{H}_s and E_i on \mathcal{H}_R . This is a very general form which applies to most systems.

$$\Rightarrow \text{Tr}_R([H_{int}, \dots]) = \text{Tr}_R\left([d_i E_i, e^{-i(t-t')(\mathcal{L}_s + \mathcal{L}_R)/\hbar} [d_l E_l, \varrho_R(0)\varrho_s(t')]]\right)$$

with $e^{-i\mathcal{L}t/\hbar} X = e^{-iHt/\hbar} X e^{iHt/\hbar}$ (see beginning of this chapter)

$$\begin{aligned} \Rightarrow e^{-i(t-t')(\mathcal{L}_s + \mathcal{L}_R)/\hbar} [d_l E_l, \varrho_R(0)\varrho_s(t')] &= \\ [d_l(t-t')E_l(t-t'), \varrho_R(0)e^{-iH_S(t-t')/\hbar}\varrho_s(t')e^{iH_S(t-t')/\hbar}] \end{aligned}$$

with

$$\begin{aligned} \hat{d}_l(t-t') &= e^{-i(t-t')H_S/\hbar} \hat{d}_l e^{i(t-t')H_S/\hbar} \\ \hat{E}_l(t-t') &= e^{-i(t-t')H_R/\hbar} \hat{E}_l e^{i(t-t')H_R/\hbar} \\ \varrho_R(t) &= \varrho_R(0) \quad \text{since } [H_R, \varrho_R(0)] = 0 \end{aligned}$$

Remark: $\hat{d}_l(-t)$ denotes the operator \hat{d}_l in interaction picture

$$\mathrm{Tr}_R([H_{int}, \dots]) = \mathrm{Tr}_R \left(\left[d_i E_i, [d_l(t-t') E_l(t-t'), \underbrace{\varrho_R(0) U_s(t-t') \varrho_s(t') U_s^\dagger(t-t')}_{=:\bar{\varrho}_s}] \right] \right)$$

with $U_s(t) := e^{-iH_S t/\hbar}$

$$\begin{aligned} \Rightarrow \mathrm{Tr}_R(\dots) &= \mathrm{Tr}_R \left(d_i E_i d_l(t-t') E_l(t-t') \varrho_R(0) \bar{\varrho}_s - d_i E_i \varrho_R(0) \bar{\varrho}_s d_l(t-t') E_l(t-t') - \right. \\ &\quad \left. - d_l(t-t') E_l(t-t') \varrho_R(0) \bar{\varrho}_s d_i E_i + \varrho_R(0) \bar{\varrho}_s d_l(t-t') E_l(t-t') d_i E_i \right) \\ &= d_i d_l(t-t') \bar{\varrho}_s \mathrm{Tr}_R(E_i E_l(t-t') \varrho_R(0)) - d_i \bar{\varrho}_s d_l(t-t') \mathrm{Tr}_R(E_i \varrho_R(0) E_l(t-t')) - \\ &\quad - d_l(t-t') \bar{\varrho}_s d_i \mathrm{Tr}_R(E_l(t-t') \varrho_R(0) E_i) + \bar{\varrho}_s d_l(t-t') d_i \mathrm{Tr}_R(\varrho_R(0) E_l(t-t') E_i) \end{aligned}$$

Define $F_{m_1 m_2}(t_1, t_2) := \mathrm{Tr}_R(E_{m_1}(t_1) E_{m_2}(t_2) \varrho_R(0)) = \langle E_{m_1}(t_1) E_{m_2}(t_2) \rangle$

$$\begin{aligned} \Rightarrow \mathrm{Tr}_R(\dots) &= d_i d_l(t-t') \bar{\varrho}_s F_{il}(0, t-t') - d_i \bar{\varrho}_s d_l(t-t') F_{li}(t-t', 0) - \\ &\quad - d_l(t-t') \bar{\varrho}_s d_i F_{il}(0, t-t') + \bar{\varrho}_s d_l(t-t') d_i F_{li}(t-t', 0) \end{aligned}$$

$$\begin{aligned} \Rightarrow i\hbar \partial_t \varrho_s(t) &= [H_S, \varrho_s(t)] - \frac{i}{\hbar} \int_0^t dt' \left(F_{il}(0, t-t') [d_i, d_l(t-t') \bar{\varrho}_s] + \right. \\ &\quad \left. F_{li}(t-t', 0) [\bar{\varrho}_s d_l(t-t'), d_i] \right) \end{aligned}$$

We now employ the widely used *Markov approximation*. Idea: The reservoir's correlation function $F_{il}(0, t-t')$ decays rapidly with $t-t'$. (The reservoir “has no memory” for its own state). For small $t-t'$ one then can approximate $\varrho_s(t')$ by

$$\varrho_s(t') \approx U_s(t'-t) \varrho_s(t) U_s^\dagger(t'-t)$$

U_s describes free propagation. One thus assumes that the system evolves freely for short a time.

$$\Rightarrow \bar{\varrho}_s = U_s(t-t') \varrho_s(t') U_s^\dagger(t-t') = \varrho_s(t)$$

$$\begin{aligned} i\hbar \partial_t \varrho_s(t) &= [H_S, \varrho_s(t)] - \frac{i}{\hbar} \int_0^t dt' \left(F_{il}(0, t-t') [d_i, d_l(t-t') \varrho_s(t)] + \right. \\ &\quad \left. F_{li}(t-t', 0) [d_l(t-t') \varrho_s(t), d_i] \right) \end{aligned}$$

Define $\tau = t - t' \quad d\tau = -dt'$

$$\Rightarrow i\hbar\partial_t \varrho_s(t) = [H_S, \varrho_s(t)] - \frac{i}{\hbar} \int_0^t d\tau \left(F_{il}(0, \tau) [d_i, d_l(\tau) \varrho_s(t)] + F_{li}(\tau, 0) [\varrho_s(t) d_l(\tau), d_i] \right)$$

Since $F_{il}(0, \tau)$ decays rapidly one can extend the integration to ∞

\Rightarrow Final result for the master equation in Markov approximation:

$$i\hbar\partial_t \varrho_s(t) = [H_S, \varrho_s(t)] - \frac{i}{\hbar} \int_0^\infty d\tau \left(F_{il}(0, \tau) [d_i, d_l(\tau) \varrho_s(t)] + F_{li}(\tau, 0) [\varrho_s(t) d_l(\tau), d_i] \right)$$

The master equation in Markov approximation

- is a linear differential equation and, in contrast to the general master equation, no integro differential equation.
- has constant coefficients (given by the integral over τ).

7.4 Spontaneous Emission

To describe spontaneous emission in free space we treat the atom as the system and the quantized radiation field as a reservoir. From the perspective of the atom, the state of the reservoir is essentially constant since spontaneously emitted photons do not return to the atom.

$$\begin{aligned} H_S &= \sum_n E_n |n\rangle \langle n|, \quad |n\rangle = \text{atomic energy levels} \\ H_R &= \int d^3k \sum_\sigma \hbar\omega_k a_{k,\sigma}^\dagger a_{k,\sigma} \\ H_{int} &= -\mathbf{d}\mathbf{E}(\mathbf{x}) = -\mathbf{d}^{(+)}\mathbf{E}^{(+)} - \mathbf{d}^{(-)}\mathbf{E}^{(-)} \end{aligned}$$

where \mathbf{x} denotes the position of the atom and we have made the rotating wave approximation.

$\mathbf{E}^{(+)}$ = positive-frequency part ($\propto a_{k,\sigma}$)

$\mathbf{d}^{(+)}$ = creation operator

- For a two-level system: $\mathbf{d}^{(+)} = \mathbf{d}_{eg} |e\rangle \langle g|$
- For two multiplets: $\mathbf{d}^{(+)} = \sum_{m_e, m_g} |J_e, m_e\rangle \langle J_g, m_g| \mathbf{d}_{m_e, m_g}$

The initial state of the radiation field is the vacuum: $\varrho_R(0) = |0\rangle\langle 0|$. In Markov approximation, the master equation becomes

$$i\hbar\dot{\varrho}_S = [H_S, \varrho_S] - \frac{i}{\hbar} \int_0^\infty d\tau \left\{ [d_i, d_j(\tau)\varrho_S] \langle E_i E_j(\tau) \rangle + [\varrho_S d_j(\tau), d_i] \langle E_j(\tau) E_i \rangle \right\} \quad (7.37)$$

Here we set ³

$$\begin{aligned} \mathbf{d}_1 &= \mathbf{d}^{(+)}; \quad \mathbf{E}_1 = \mathbf{E}^{(+)} \\ \mathbf{d}_2 &= \mathbf{d}^{(-)}; \quad \mathbf{E}_2 = \mathbf{E}^{(-)} \end{aligned}$$

It is easy to see that for the vacuum state for the photons one has

$$\langle 0 | E_a^{(+)} E_b^{(+)} | 0 \rangle = \langle 0 | E_a^{(-)} E_b^{(-)} | 0 \rangle = \langle 0 | E_a^{(-)} E_b^{(+)} | 0 \rangle = 0,$$

resulting in the master equation

$$\begin{aligned} \left\{ [d_i, \dots] \langle \dots \rangle \dots \right\} &= \left\{ \langle E_a^{(+)} E_b^{(-)}(\tau) \rangle [d_a^{(+)}, d_b^{(-)}(\tau)\varrho_S] \right. \\ &\quad \left. + \langle E_b^{(+)}(\tau) E_a^{(-)} \rangle [\varrho_S d_b^{(+)}(\tau), d_a^{(-)}] \right\} \end{aligned}$$

The correlation function has the following form (the derivation is left as an exercise for the reader)

$$\langle 0 | E_a^{(+)}(\mathbf{x}, 0) E_b^{(-)}(\mathbf{x}, \tau) | 0 \rangle = \frac{\hbar}{6\pi^2 \epsilon_0 c^3} \delta_{ab} \int_0^\infty d\omega_k \omega_k^3 e^{-i\omega_k \tau} \quad (7.38)$$

With the aid of the free evolution operator $U(\tau)$, the second correlation function, in which τ appears in the first operator, can be expressed as

$$\langle 0 | E_a^{(+)}(\tau) E_b^{(-)}(0) | 0 \rangle = \langle 0 | U(\tau) E_a^{(+)}(0) U^\dagger(\tau) E_b^{(-)}(0) | 0 \rangle$$

Use $U | 0 \rangle = | 0 \rangle$:

$$\begin{aligned} \Rightarrow \langle 0 | E_a^{(+)}(\tau) E_b^{(-)}(0) | 0 \rangle &= \langle 0 | E_a^{(+)}(0) U^\dagger(\tau) E_b^{(-)}(0) U(\tau) | 0 \rangle \\ &= \langle 0 | E_a^{(+)}(0) E_b^{(-)}(-\tau) | 0 \rangle \end{aligned}$$

The time dependence of the atomic operators for free evolution is given by

$$\mathbf{d}^{(+)}(\tau) = \sum_{m_e, m_g} |J_e m_e\rangle \langle J_g m_g| \mathbf{d}_{m_e, m_g} e^{i\tau(E_{m_g} - E_{m_e})/\hbar}$$

³Strictly speaking each d_i in the sum above denotes a vector component of either $\mathbf{d}^{(+)}$ or $\mathbf{d}^{(-)}$. To keep the notation concise we omit this here.

$$\begin{aligned} \Rightarrow & \int_0^\infty d\tau d_a^{(+)}(\tau) \langle E_a^{(+)}(\tau) E_b^{(-)}(0) \rangle \\ &= \frac{\hbar}{6\pi^2\epsilon_0 c^3} \sum_{m_e, m_g} (\mathbf{d}_{m_e, m_g})_b |J_e, m_e\rangle \langle J_g, m_g| \int_0^\infty \omega^3 d\omega \int_0^\infty d\tau e^{i\tau(\omega - \omega_{m_e, m_g})} \end{aligned}$$

with $\omega_{m_e, m_g} := (E_{m_e} - E_{m_g})/\hbar$. Using $\int_0^\infty d\tau e^{-i\omega\tau} = \pi\delta(\omega) - i\mathcal{P}/\omega$ one arrives at

$$\begin{aligned} \int_0^\infty d\tau d_a^{(+)}(\tau) \langle E_a^{(+)}(\tau) E_b^{(-)}(0) \rangle &= \frac{\hbar}{6\pi^2\epsilon_0 c^3} \sum_{m_e, m_g} |J_e, m_e\rangle \langle J_g, m_g| (\mathbf{d}_{m_e, m_g})_b \\ &\quad \times \left(\pi\omega_{m_e, m_g}^3 + i \int_0^\infty d\omega \frac{\mathcal{P}\omega^3}{(\omega - \omega_{m_e, m_g})} \right) \\ &=: \sum_{m_e, m_g} |J_e, m_e\rangle \langle J_g, m_g| (\mathbf{d}_{m_e, m_g})_b (K_{m_e m_g} + iK'_{m_e m_g}) \\ &=: \hat{\mathbf{K}}_b^{(+)} + i\hat{\mathbf{K}}_b'^{(+)} \end{aligned}$$

The operators $\hat{\mathbf{K}}^{(+)}$ and $\hat{\mathbf{K}}'^{(+)}$ only act on the atomic degrees of freedom and are time independent. Our result for the master equation for atomic multiplets then becomes

$$\boxed{i\hbar\dot{\varrho}_S = [H_S, \varrho_S] - \frac{i}{\hbar} \left\{ [d_a^{(+)}, (\hat{K}_a^{(-)} - i\hat{K}_a'^{(-)})\varrho_S] + [\varrho_S(\hat{K}_a^{(+)} + i\hat{K}_a'^{(+)}), d_a^{(-)}] \right\}} \quad (7.39)$$

where we have introduced $\hat{\mathbf{K}}^{(-)} := (\hat{\mathbf{K}}^{(+)})^\dagger$ and $\hat{\mathbf{K}}'^{(-)} := (\hat{\mathbf{K}}'^{(+)})^\dagger$.

For a two-level system these operators have the simple form

$$\hat{\mathbf{K}}^{(+)} = |e\rangle\langle g| \mathbf{d}_{eg} \frac{\hbar\omega_0^3}{6\pi\epsilon_0 c^3}; \quad \hat{\mathbf{K}}'^{(+)} = |e\rangle\langle g| \mathbf{d}_{eg} \int_0^\infty \dots \quad (7.40)$$

The numerical value of the integral in $\hat{\mathbf{K}}'^{(+)}$ is irrelevant. It not only diverges (and therefore has to be renormalized), but it also leads to an incorrect value when it is calculated in two-level approximation and rotating wave approximation. Inserting these operators into Eq. (7.39) we find the *Master equation for two-level atoms*

$$\begin{aligned} \dot{\varrho} &= -\frac{i}{\hbar} [\hbar\omega_0 |e\rangle\langle e|, \varrho] - \frac{i}{\hbar} [\hbar\Delta_{\text{Lamb}} |e\rangle\langle e|, \varrho] \\ &\quad - \frac{\gamma}{2} (\sigma_+ \sigma_- \varrho + \varrho \sigma_+ \sigma_- - 2\sigma_- \varrho \sigma_+) \end{aligned} \quad (7.41)$$

with

$$\gamma := \frac{|\mathbf{d}_{eg}|^2 \omega_0^3}{3\pi \hbar \epsilon_0 c^3} \quad (7.42)$$

and

$$\Delta_{\text{Lamb}} := -\frac{|\mathbf{d}_{eg}|^2}{6\pi^2 \hbar \epsilon_0 c^3} \int_0^\infty d\omega \frac{\mathcal{P}\omega^3}{(\omega - \omega_0)}$$

Consequences Obviously, the real part $\mathbf{K}^{(+)}$ and the imaginary part $\mathbf{K}'^{(+)}$ describe very different phenomena.

- $\hat{\mathbf{K}}'^{(+)}$ contains an integral over ω with a principal value and is therefore related to the commutator $\propto \Delta_L$. It describes a shift in the resonance frequency: $\omega'_0 = \omega_0 + \Delta_L$. Δ_L is called *Lamb shift*.
- Δ_L is divergent and needs to be renormalized. A derivation which goes beyond the two-level approximation can be found in Ref. [8]. Δ_L does not change the unitarity of the time evolution.
- The real part $\hat{\mathbf{K}}^{(+)}$ is the origin for the term proportional to γ which describes a non-unitary time evolution (see below); γ denotes the *spontaneous emission rate*.
- The expression $-\frac{\gamma}{2}(R^\dagger R \varrho + \varrho R^\dagger R - 2R \varrho R^\dagger)$ corresponds to the most general form of an incoherent time evolution which conserves the probability and is local in time. It is called Lindblad form. R is a generalized raising or lowering operator.

Example: a thermal radiation reservoir at temperature T instead of $|0\rangle \implies$ thermal excitation of the atom:

$$-\eta(T)(\sigma_- \sigma_+ \varrho + \varrho \sigma_- \sigma_+ - 2\sigma_+ \varrho \sigma_-) .$$

More on this can be found in Ref. [6].

In the following we will assume that the resonance frequency ω_0 already contains the Lamb shift, $\omega_0 \equiv \omega'_0 + \Delta_L$. The master equation is given by

$$\dot{\varrho} = -\frac{i}{\hbar}[\hbar\omega_0 |e\rangle\langle e|, \varrho] - \frac{\gamma}{2}\{\sigma_+ \sigma_- \varrho + \varrho \sigma_+ \sigma_- - 2\sigma_- \varrho \sigma_+\} \quad (7.43)$$

We write the density matrix as

$$\varrho = \begin{pmatrix} \varrho_{ee} & \varrho_{eg} \\ \varrho_{ge} & \varrho_{gg} \end{pmatrix}$$

and first calculate the commutator with the system Hamiltonian:

$$[|e\rangle\langle e|, \varrho] = \left[\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \varrho \right] = \begin{pmatrix} 0 & \varrho_{eg} \\ -\varrho_{ge} & 0 \end{pmatrix}$$

For the Lindblad form we find

$$\sigma_+ \sigma_- = |e\rangle\langle g|g\rangle\langle e| = |e\rangle\langle e|$$

so that

$$|e\rangle\langle e| \varrho + \varrho |e\rangle\langle e| - 2 \underbrace{|g\rangle\langle e|}_{\sigma_-} \varrho \underbrace{|e\rangle\langle g|}_{\sigma_+} = \begin{pmatrix} 2\varrho_{ee} & \varrho_{eg} \\ \varrho_{ge} & -2\varrho_{ee} \end{pmatrix}$$

The master equation then can be brought into the form

$$\begin{pmatrix} \dot{\varrho}_{ee} & \dot{\varrho}_{eg} \\ \dot{\varrho}_{ge} & \dot{\varrho}_{gg} \end{pmatrix} = -i\omega_0 \begin{pmatrix} 0 & \varrho_{eg} \\ -\varrho_{ge} & 0 \end{pmatrix} - \frac{\gamma}{2} \begin{pmatrix} 2\varrho_{ee} & \varrho_{eg} \\ \varrho_{ge} & -2\varrho_{ee} \end{pmatrix} \quad (7.44)$$

which results in the following equations for the matrix components:

$$\begin{aligned} \dot{\varrho}_{ee} &= -\gamma\varrho_{ee} & \Rightarrow & \varrho_{ee}(t) = \varrho_{ee}(0)e^{-\gamma t} \\ \dot{\varrho}_{eg} &= \left(-i\omega_0 - \frac{\gamma}{2}\right) \varrho_{eg} & \Rightarrow & \varrho_{eg}(t) = e^{-i\omega_0 t} e^{-\frac{\gamma}{2}t} \varrho_{eg}(0) \\ \dot{\varrho}_{gg} &= \gamma\varrho_{ee} & \Rightarrow & \varrho_{gg}(t) = \varrho_{gg}(0) + (1 - e^{-\gamma t})\varrho_{ee}(0) \end{aligned} \quad (7.45)$$

This solution is characteristic for spontaneous emission:

- The excited state decays incoherently since during the transition from e to g no coherences ϱ_{eg} , ϱ_{ge} do appear. In the case of coherent Rabi oscillations one finds that ϱ_{eg} , ϱ_{ge} first start to grow.
- If there are initial coherences ϱ_{eg} , ϱ_{ge} they decay with rate $\frac{\gamma}{2}$. This is because $|g\rangle\langle e|$ contains an excited component which can decay. $|e\rangle\langle e|$, on the other hand, contains two excited components.
- The population lost in ϱ_{ee} is transferred to ϱ_{gg} , so that probability is conserved.

$$\text{Tr } \varrho = 1 \quad \rightarrow \quad \text{Tr } \dot{\varrho} = 0 = \dot{\varrho}_{ee} + \dot{\varrho}_{gg}$$

This is a general feature of the Lindblad form:

$$\begin{aligned} \text{Tr } \dot{\varrho} &= -\frac{\gamma}{2} \text{Tr} \{ R^\dagger R \varrho + \varrho R^\dagger R - 2R \varrho R^\dagger \} \\ &= -\frac{\gamma}{2} \text{Tr} \{ R^\dagger R \varrho + R^\dagger R \varrho - 2R^\dagger R \varrho \} = 0 \end{aligned}$$

- The master equation for the density matrix is superior to a phenomenological description of spontaneous emission based on the wavefunction only. With $\psi_e(t) = e^{-\frac{\gamma}{2}t}\psi_e(0)$ We find for the trace of ϱ :

$$|\psi_e|^2 + |\psi_g|^2 = e^{-\gamma t}|\psi_e(0)|^2 + |\psi_g(0)|^2 \neq \text{const.}$$

Probability is therefore not conserved. One may try to repair this by some ad hoc ansatz, but such an approach leads to the creation of coherences, which should not occur in an incoherent process like spontaneous emission.

$$\psi_g(t) \propto \sqrt{|\psi_g(0)|^2 + (1 - e^{-\gamma t})|\psi_e(0)|^2} \Rightarrow \varrho_{eg} = \psi_e \psi_g^* \neq 0$$

For $t = 0$ and $t \rightarrow \infty$ the coherence ϱ_{eg} is going to zero, but during the evolution this is not the case.

Visualization on the Bloch sphere: see Fig. 7.1

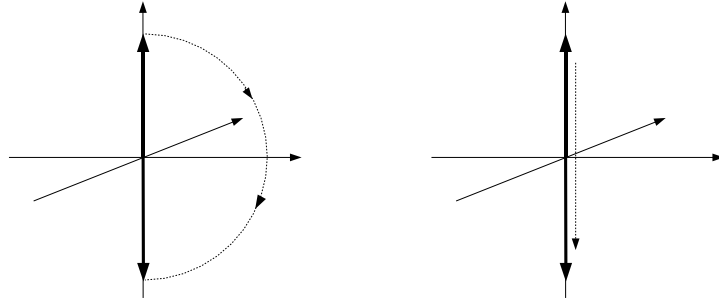


Figure 7.1: π pulse (left) and spontaneous emission (right) on the Bloch sphere.

Both for a Rabi oscillation with total angle π and for spontaneous emission the initial and final state are the same. The difference becomes evident on the Bloch sphere: for Rabi oscillations, the Bloch vector moves on a semi-circle its length remains unchanged. The system always remains in a pure state. During spontaneous emission, the Bloch vector becomes smaller and evolves right through the center of the Bloch sphere, which corresponds to a maximally mixed state.

Let us now consider the case when the initial state is an arbitrary superposition. Without spontaneous emission the Bloch vector evolves on a circle on the surface of the sphere (Fig. 7.2, In the presence of spontaneous emission, this is changed to a spiral downwards which ends in the ground state. Again the state is a mixed state for intermediate times (Fig. 7.2, right). To demonstrate the transition from a pure state to a superposition we calculate the trace of ϱ^2 :

$$\text{Tr } \varrho^2 = \varrho_{ee}^2 + 2\varrho_{eg}\varrho_{ge} + \varrho_{gg}^2$$

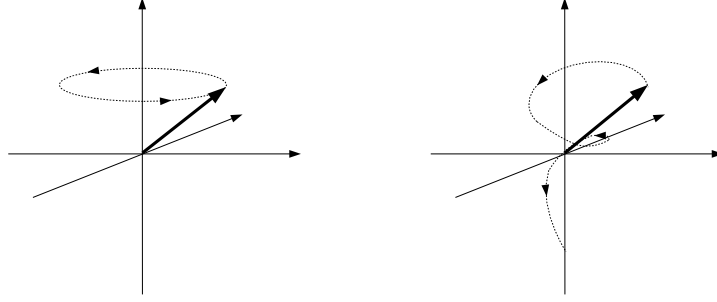


Figure 7.2: Evolution of an arbitrary superposition for a Rabi oscillation (left) and for spontaneous emission (right).

For $\varrho_{ee}(0) = 1$ we find from Eq. (7.45) the result

$$\text{Tr } \varrho^2(t) = 1 - 2e^{-\gamma t} + 2e^{-2\gamma t} \quad (7.46)$$

The time evolution is shown in Fig. Fig. 7.3. The absolute value of $\text{Tr } \varrho^2$ quickly

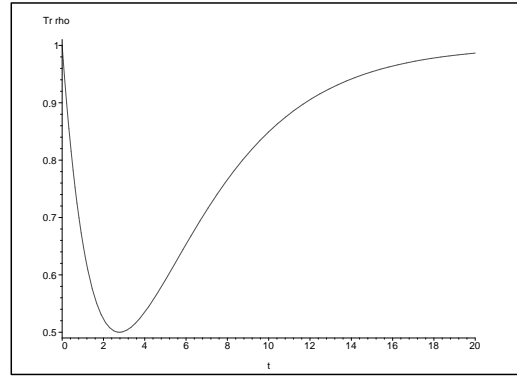


Figure 7.3: $\text{Tr } \varrho^2$ during spontaneous emission

drops to the value of $\frac{1}{2}$ (complete mixture) and then asymptotically returns to one again. This return to a pure state is only present if one neglects the atomic center-of-mass motion. Because of the momentum transfer associated with a spontaneously emitted photon the final state of spontaneous emission corresponds to a mixture in momentum space. This process can roughly be described as

$$\begin{aligned} \varrho &= |e, \mathbf{p}\rangle\langle e, \mathbf{p}| \rightarrow |e, \mathbf{p}\rangle\langle e, \mathbf{p}| + \int d^3k |g, \mathbf{p} + \hbar\mathbf{k}\rangle\langle g, \mathbf{p} + \hbar\mathbf{k}| \delta(|\mathbf{k}| - \frac{\omega_0}{c}) \\ &\rightarrow \int d^3k |g, \mathbf{p} + \hbar\mathbf{k}\rangle\langle g, \mathbf{p} + \hbar\mathbf{k}| \delta(|\mathbf{k}| - \frac{\omega_0}{c}) \end{aligned} \quad (7.47)$$

7.5 Spontaneous emission cancellation through interference

We consider a three-level atom in V-configuration:

$$\begin{array}{c}
 \begin{array}{cc}
 |+\rangle & |-\rangle \\
 \hline
 \end{array} \\
 \hline
 |g\rangle \\
 E_e = E_+ = E_-
 \end{array}$$

In the general master equation (7.39) for atoms one then finds a term of the form

$$\begin{aligned}
 \mathbf{K}_i^{(+)} + i\mathbf{K}_i'^{(+)} &= \int_0^\infty d\tau \mathbf{d}_j^{(+)}(\tau) \langle 0 | E_j^{(+)}(\tau) E_i^{(-)}(\tau) | 0 \rangle \\
 &= \sum_{m_e=\pm} |m_e\rangle \langle g| (\mathbf{d}_{m_e g})_i \underbrace{(K_{m_e g})}_{\in \mathbb{R}} + i \underbrace{K'_{m_e g}}_{\in \mathbb{R}}
 \end{aligned}$$

Since $K_{m_e g}$, $K'_{m_e g}$ only depend on $E_e - E_g$ they are equal for both transitions, $+\rightarrow g$ and $-\rightarrow g$.

$$\mathbf{K}^{(+)} + i\mathbf{K}'^{(+)} = (K + iK') \{ |+\rangle \langle g| \mathbf{d}_{+g} + |-\rangle \langle g| \mathbf{d}_{-g} \}$$

Inserting this into the master equation leads to

$$\begin{aligned}
 i\hbar \dot{\varrho}_s &= [H_S, \varrho_s] \\
 &- \frac{i}{\hbar} \left\{ \left[\mathbf{d}_{+g} |+\rangle \langle g| + \mathbf{d}_{-g} |-\rangle \langle g| \ , \ (K - iK') (|g\rangle \langle +| \mathbf{d}_{+g}^* + |g\rangle \langle -| \mathbf{d}_{-g}^*) \varrho_s \right] \right. \\
 &\left. + (K + iK') \left[\varrho_s (|+\rangle \langle g| \mathbf{d}_{+g} + |-\rangle \langle g| \mathbf{d}_{-g}) \ , \ \mathbf{d}_{+g}^* |g\rangle \langle +| + \mathbf{d}_{-g}^* |g\rangle \langle -| \right] \right\}
 \end{aligned}$$

Under the *assumption* that ⁴ $\mathbf{d}_+ = \mathbf{d}_- = \mathbf{d}$ this can be reduced to

$$\begin{aligned}
 i\hbar \dot{\varrho}_s &= [H_S, \varrho_s] - \frac{i}{\hbar} |\mathbf{d}|^2 \left\{ (K - iK') \left[(|+\rangle + |-\rangle) \langle g| \ , \ |g\rangle (\langle +| + \langle -|) \varrho_s \right] \right. \\
 &\left. + (K + iK') \left[\varrho_s (|+\rangle + |-\rangle) \langle g| \ , \ |g\rangle (\langle +| + \langle -|) \right] \right\}
 \end{aligned} \tag{7.48}$$

⁴The choice $\mathbf{d}_+ = e^{i\alpha} \mathbf{d}_-$ is also possible.

If the atoms initially are in the special excited state $|\psi\rangle = \frac{1}{\sqrt{2}}(|+\rangle - |-\rangle)$ we find

$$\varrho_s(0) = |\psi\rangle\langle\psi|, \quad H_S |\psi\rangle = E_e |\psi\rangle \rightarrow [H_S, \varrho_s(0)] = 0$$

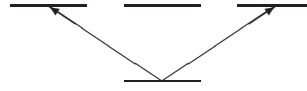
In addition we have $(\langle+| + \langle-|) |\psi\rangle = 0$. Because of the term $\varrho_s(|+\rangle + |-\rangle) = 0$ that appears in the commutators in Eq. (7.48) one then finds

$$\dot{\varrho}_s = 0$$

$\Rightarrow |\psi\rangle$ can not decay into the ground state!

- Destructive interference between the decay amplitudes of $|+\rangle$ and $|-\rangle$ causes this superposition to be stable [14].
- This effect reminds us of the dark state in a Λ -system. The major difference is that $|\psi\rangle$ is an excited state and represents a dark state for all resonant photons.
- The observation of this effect is difficult since the assumption $\mathbf{d}_+ = e^{i\alpha} \mathbf{d}_-$ cannot be fulfilled for atoms. This is because atomic energy eigenstates are eigenstates of J_z

Example: three-level system in V configuration:



$$J_g = 0 \quad J_e = 1$$

One then has

$$\begin{aligned} \mathbf{d}_+ &= \mathcal{D}(\mathbf{e}_x - i\mathbf{e}_y)/\sqrt{2} && \text{couples to } \sigma_+ \text{ light} \\ \mathbf{d}_- &= \mathcal{D}(\mathbf{e}_x + i\mathbf{e}_y)/\sqrt{2} && \text{couples to } \sigma_- \text{ light} \end{aligned}$$

Thus, $\mathbf{d}_+ \cdot \mathbf{d}_-^* = 0$ and the interference terms in the master equation are zero. For instance, we have

$$(|+\rangle \mathbf{d}_+ + |-\rangle \mathbf{d}_-) \langle g| \cdot |g\rangle (\langle+| \mathbf{d}_+^* + \langle-| \mathbf{d}_-^*) = \mathcal{D}^2(|+\rangle\langle+| + |-\rangle\langle-|).$$

By contrast, under the assumption $\mathbf{d}_+ = \mathbf{d}_-$ used above one finds

$$\mathcal{D}^2(|+\rangle + |-\rangle)(\langle+| + \langle-|).$$

- For molecules destructive interference is possible in principle. Experimental observation of spontaneous emission cancellation was reported in Ref. [15], but the result was later questioned [16].

Chapter 8

Electromagnetically induced transparency

EIT is one of many effects in quantum optics where light changes the state of the atoms, which in turn leads to a change of the index of refraction. The description of such effects is based on

8.1 The Maxwell-Bloch equations

Equation for atoms: $\dot{\varrho} = \mathcal{L}\varrho$ as in Chapter 7, with the interaction between light and atoms given by

$$\begin{aligned} H &= H_A + H_{\text{Rad}} - \frac{1}{\epsilon_0} \int \mathbf{P}(\mathbf{x}) \cdot \mathbf{D}(\mathbf{x}) d^3x \\ \mathbf{P}(\mathbf{x}) &= \text{Polarization; e.g. for a single atom at position } \mathbf{x}_0 : \\ \mathbf{P}(\mathbf{x}) &= \hat{\mathbf{d}}\delta(\mathbf{x} - \mathbf{x}_0) \end{aligned} \quad (8.1)$$

Heisenberg's equation of motion: (as in Chapter 2):

$$\begin{aligned} i\hbar \dot{\mathbf{D}}(\mathbf{x}) &= [\mathbf{D}(\mathbf{x}), H] \text{ with} \\ [D_i(\mathbf{x}), B_j(\mathbf{x}')] &= -i\hbar \epsilon_{ijk} \partial_k \delta(\mathbf{x} - \mathbf{x}') \end{aligned} \quad (8.2)$$

from which follows

$$\dot{\mathbf{D}} = \frac{1}{\mu_0} \text{curl } \mathbf{B} \quad (8.3)$$

$$\dot{\mathbf{B}} = -\frac{1}{\epsilon_0} \text{curl } (\mathbf{D} - \mathbf{P}) \quad (8.4)$$

These are Maxwell's equations ¹ in a dielectric with polarization \mathbf{P} . As we have learned in the first chapters, one can deduce wave equations from these relations:

$$\left(\frac{1}{c^2} \partial_t^2 + \text{curl curl} \right) \mathbf{D} = \text{curl curl} \mathbf{P}, \quad (8.5)$$

or, using $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$,

$$\left(\frac{1}{c^2} \partial_t^2 + \text{curl curl} \right) \mathbf{E} = -\mu_0 \ddot{\mathbf{P}} \quad (8.6)$$

Together with $\dot{\varrho} = \mathcal{L}\varrho$ (see Chapter 7) this equation represents the *Maxwell-Bloch equations*. These equations are generally hard to solve and one has to make approximations. Frequently used assumptions are the following.

- Homogeneously distributed atoms

$$\Rightarrow \mathbf{P}(\mathbf{x}, t) = \mathbf{P}(t) \quad (8.7)$$

- The atomic density is very low so that the atoms do not interact directly with each other (collisions) and do only interact with the light.

$$\Rightarrow \mathbf{P}(t) = \bar{\varrho} \mathbf{d} \quad (8.8)$$

here $\bar{\varrho}$ denotes the atomic density and \mathbf{d} the dipole moment operator for a single atom.

Averaging over the atomic degrees-of-freedom results in

$$\left(\frac{1}{c^2} \partial_t^2 + \text{curl curl} \right) \mathbf{E} = -\mu_0 \bar{\varrho} \partial_t^2 \langle \mathbf{d} \rangle \quad (8.9)$$

$$= -\frac{1}{c^2 \epsilon_0} \bar{\varrho} \text{Tr}(\ddot{\varrho} \mathbf{d}) \quad (8.10)$$

The Maxwell-Bloch equation allow to calculate the optical properties of an atomic medium. In a gas one can neglect interatomic collisions to a certain degree.

¹We assume that the macroscopic free charge density vanishes, so that $\text{div} \mathbf{D} = 0$. It is then not necessary to distinguish between the transverse part of the dielectric displacement field and the field itself.

8.2 Index of refraction for a two-level atom

We consider a two-level atom in a running laser beam along the z -axis,

$$\mathbf{E} = \mathcal{E}(z)\boldsymbol{\epsilon}e^{ik_L z - i\omega_L t} + \text{c.c.} \quad (8.11)$$

The laser beam is continuously switched on (“cw”) so that the envelope $\mathcal{E}(\mathbf{x})$ is time independent. For simplicity we assume that the width of the laser beam is much larger than the wavelength $2\pi/k_L$ so that the envelope depends on z only. If the variation of the envelope is small on the scale of the wavelength, we can make use of the paraxial approximation (see Sec. 1.8). The wave equation (8.9) is then reduced to

$$2ik_L \boldsymbol{\epsilon} e^{ik_L z - i\omega_L t} \partial_z \mathcal{E} - 2ik_L \boldsymbol{\epsilon}^* e^{-ik_L z + i\omega_L t} \partial_z \mathcal{E}^* = -\frac{1}{c^2 \epsilon_0} \bar{\varrho} \text{Tr}(\ddot{\mathbf{d}}) \quad (8.12)$$

Our task is to find a suitable approximation for the atomic density matrix ϱ so that we can solve this equation.

8.2.1 Stationary solution for a driven two-level atom

The evolution of the atomic density matrix is governed by the master equation. For a two-level atom in free space it is given by (7.43). In the presence of a laser beam, we have to replace the atomic Hamiltonian $\hbar\omega_0 |e\rangle\langle e|$ by $H = \hbar\omega_0 |e\rangle\langle e| - \mathbf{d} \cdot \mathbf{E}$. Proceeding as in Chapter 6 we find in the rotating-wave approximation the Hamiltonian (6.17), i.e.,

$$H = \hbar\omega_0 |e\rangle\langle e| - \hbar\Omega e^{-i\omega_L t} |e\rangle\langle g| - \hbar\Omega^* e^{i\omega_L t} |g\rangle\langle e| \quad (8.13)$$

where

$$\Omega \equiv \frac{\mathbf{d}_{eg} \cdot \mathbf{E}^{(+)}(t=0)}{\hbar} = \frac{\mathbf{d}_{eg} \cdot \boldsymbol{\epsilon} e^{ik_L z} \mathcal{E}}{\hbar} \quad (8.14)$$

Similarly as in Sec. 6.2 we can make a unitary transformation

$$U(t) = \exp(-i\omega_L t |e\rangle\langle e|) \quad (8.15)$$

With $\varrho = U \tilde{\varrho} U^\dagger$ we find

$$\begin{aligned} \tilde{H} &= -i\hbar U^\dagger \dot{U} + U^\dagger H U \\ &= -\hbar\Delta |e\rangle\langle e| - \hbar\Omega |e\rangle\langle g| - \hbar\Omega^* |g\rangle\langle e| \end{aligned} \quad (8.16)$$

with $\Delta \equiv \omega_L - \omega_0$, so that

$$\dot{\tilde{\varrho}} = -\frac{i}{\hbar} [\tilde{H}, \tilde{\varrho}] - \frac{\gamma}{2} \{ |e\rangle\langle e| \tilde{\varrho} + \tilde{\varrho} |e\rangle\langle e| - 2 |g\rangle\langle e| \tilde{\varrho} |e\rangle\langle g| \} \quad (8.17)$$

This is a set of linear differential equations with constant coefficients. If $\tilde{\rho}$ is written as

$$\tilde{\rho} = \tilde{\rho}_{ee} |e\rangle\langle e| + \tilde{\rho}_{eg} |e\rangle\langle g| + \tilde{\rho}_{ge} |g\rangle\langle e| + \tilde{\rho}_{gg} |g\rangle\langle g| \quad (8.18)$$

the equations become

$$\begin{aligned} \dot{\tilde{\rho}}_{ee} &= -\gamma \tilde{\rho}_{ee} - i(\tilde{\rho}_{ge} \Omega - \tilde{\rho}_{eg} \Omega^*) \\ \dot{\tilde{\rho}}_{eg} &= \left(i\Delta - \frac{\gamma}{2}\right) \tilde{\rho}_{eg} + i\Omega(\tilde{\rho}_{ee} - \tilde{\rho}_{gg}) . \end{aligned} \quad (8.19)$$

The equations for $\tilde{\rho}_{ge}$ and $\tilde{\rho}_{gg}$ are not needed because of $\tilde{\rho}_{ge} = \tilde{\rho}_{eg}^*$ and $\text{Tr}(\tilde{\rho}) = \tilde{\rho}_{ee} + \tilde{\rho}_{gg} = 1$.

To consider a cw laser field has a number of advantages. After a sufficiently large time (large compared to γ^{-1}) all transient excitations will be damped away by spontaneous emission. The atoms then settle in a stationary state in which coherent (deexcitation and) excitation by the laser field and incoherent deexcitation cancel each other. This stationary state is determined by setting $\dot{\tilde{\rho}} = 0$, leading to

$$\begin{aligned} \tilde{\rho}_{ee}^{st} &= \frac{|\Omega|^2}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega|^2} \\ \tilde{\rho}_{gg}^{st} &= 1 - \tilde{\rho}_{ee}^{st} \\ \tilde{\rho}_{eg}^{st} &= \frac{\Omega(\Delta - i\frac{\gamma}{2})}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega|^2} . \end{aligned} \quad (8.20)$$

This is the solution in the “co-rotating frame”. To get the solution in the “laboratory frame”, we need to undo the unitary transformation $U = \exp(-i\omega_L t |e\rangle\langle e|)$. It follows that

$$\begin{aligned} \rho(t) &= U(t) \tilde{\rho}^{st} U^\dagger(t) \\ &= \sum_{i,j} \tilde{\rho}_{ij}^{st} U |i\rangle\langle j| U^\dagger \\ &= \tilde{\rho}_{ee}^{st} |e\rangle\langle e| + \tilde{\rho}_{gg}^{st} |g\rangle\langle g| + \tilde{\rho}_{eg}^{st} e^{-i\omega_L t} |e\rangle\langle g| + \tilde{\rho}_{ge}^{st} e^{i\omega_L t} |g\rangle\langle e| \end{aligned} \quad (8.21)$$

8.2.2 Solution of the paraxial wave equation

We now employ this solution to calculate the right-hand side of (8.12) by using

$$\text{Tr}(\rho(t) \mathbf{d}) = \mathbf{d}_{eg} \rho_{ge}(t) + \mathbf{d}_{eg}^* \rho_{eg}(t) . \quad (8.22)$$

Inserting this into Maxwell’s equation results in

$$\epsilon e^{ik_L z - i\omega_L t} \partial_z \mathcal{E} - \epsilon^* e^{-ik_L z + i\omega_L t} \partial_z \mathcal{E}^* = -i \frac{k_L \bar{\rho}}{2\epsilon_0} (\mathbf{d}_{eg} \tilde{\rho}_{ge}^{st} e^{i\omega_L t} + \mathbf{d}_{eg}^* \tilde{\rho}_{eg}^{st} e^{-i\omega_L t}) \quad (8.23)$$

Comparing the rapidly oscillating exponential factors (and polarizations) one thus finds

$$\begin{aligned}
 \epsilon e^{ik_L z} \partial_z \mathcal{E} &= -i \frac{k_L \bar{\rho}}{2\epsilon_0} \mathbf{d}_{eg}^* \tilde{\rho}_{eg}^{st} \\
 &= -i \frac{k_L \bar{\rho}}{2\epsilon_0} \mathbf{d}_{eg}^* \frac{\Omega(\Delta - i\frac{\gamma}{2})}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega|^2} \\
 &= -i \frac{k_L \bar{\rho}}{2\epsilon_0 \hbar} \mathbf{d}_{eg}^* \frac{\Delta - i\frac{\gamma}{2}}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega|^2} \mathbf{d}_{eg} \cdot \epsilon e^{ik_L z} \mathcal{E} \quad (8.24)
 \end{aligned}$$

From the Wigner-Eckart theorem (5.78) we know that

$$\begin{aligned}
 \mathbf{d}_{eg} &= \langle F_e, m_e | \mathbf{d} | F_g, m_g \rangle \\
 &= \langle F_e, m_e | d_q | F_g, m_g \rangle \epsilon_q^* \\
 &= \langle F_g, m_g; 1, q | F_e, m_e \rangle \epsilon_q^* \frac{\mathcal{D}(F_e, F_g)}{\sqrt{2F_e + 1}} \quad (8.25)
 \end{aligned}$$

If the laser beam is either linearly or circularly² polarized, we can choose the quantization axis in such a way that one vector ϵ_{q_0} coincides with the laser polarization ϵ . Hence,

$$\mathbf{d}_{eg} = \mathcal{D} \epsilon^* \quad (8.26)$$

with $\mathcal{D} \equiv \langle F_g, m_g; 1, q_0 | F_e, m_e \rangle \mathcal{D}(F_e, F_g) / \sqrt{2F_e + 1}$. The paraxial equation thus reduces to

$$\partial_z \mathcal{E} = -i \frac{k_L \bar{\rho}}{2\epsilon_0 \hbar} |\mathcal{D}|^2 \frac{\Delta - i\frac{\gamma}{2}}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega|^2} \mathcal{E} \quad (8.27)$$

In this chapter we make the additional assumption that the factor of $|\Omega|^2$ in the denominator can be kept constant. One then arrives at a simple linear equation for \mathcal{E} with the solution

$$\mathcal{E}(z) = \exp \left(-iz \frac{k_L |\mathcal{D}|^2 \bar{\rho}}{2\epsilon_0 \hbar} \frac{\Delta - i\frac{\gamma}{2}}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega_0|^2} \right) \mathcal{E}(0) \quad (8.28)$$

For the complete positive-frequency part of the electric field this implies

$$\mathbf{E}^{(+)} = \epsilon \exp \left\{ -i\omega_L t + ik_L z \left(1 - \frac{|\mathcal{D}|^2 \bar{\rho}}{2\epsilon_0 \hbar} \frac{\Delta - i\frac{\gamma}{2}}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega_0|^2} \right) \right\} \mathcal{E} \quad (8.29)$$

²This excludes elliptically polarized light, but it seems not be possible to construct a real two-level system for this case anyway.

It then becomes obvious that the solution of the Maxwell-Bloch equations corresponds to a (complex) change in the wavevector of the electric field. We can compare this change with the definition of the index of refraction :

$$k(\omega_L) = \frac{\omega_L}{c} n(\omega_L) \quad (8.30)$$

leading to

$$n(\omega_L) = 1 - \frac{\bar{\rho}|\mathcal{D}|^2}{2\hbar\epsilon_0} \frac{\Delta - i\frac{\gamma}{2}}{\Delta^2 + \frac{\gamma^2}{4} + 2|\Omega_0|^2} \quad (8.31)$$

The index of refraction is generally complex, $n = n' + in''$

$$\begin{aligned} \Rightarrow \mathbf{E}^{(+)} &= \mathbf{E}_0 e^{-i\omega_L t} e^{i\mathbf{k}x \frac{\omega_L}{c} (n' + in'')} \\ &= \mathbf{E}_0 e^{-i\omega_L t} e^{i\mathbf{k}'x} e^{-\mathbf{k}''x} \end{aligned} \quad (8.32)$$

- The real part n' determines the phase velocity $v_{ph} = \frac{\omega}{\text{Re } k}$ of the light. For vanishing detuning the phase velocity becomes $v(\Delta = 0) = c$.
- Group velocity: Consider a wavepacket

$$E(x, t) = \int d\omega e^{ik(\omega)x} e^{-i\omega t} E(\omega) \quad (8.33)$$

For a narrow frequency width $\omega \approx \omega_0$, we can make the expansion

$$k(\omega) \approx k(\omega_0) + \left. \frac{\partial k}{\partial \omega} \right|_{\omega_0} (\omega - \omega_0) + \dots \quad (8.34)$$

$$\begin{aligned} \Rightarrow E(x, t) &= \int d\omega E(\omega) e^{-i\omega t} e^{ik(\omega_0)x} e^{i \left. \frac{\partial k}{\partial \omega} \right|_{\omega_0} (\omega - \omega_0)x} \\ &= e^{ik(\omega_0)x} e^{-i \left. \frac{\partial k}{\partial \omega} \right|_{\omega_0} \omega_0 x} \int d\omega E(\omega) e^{-i\omega(t - \left. \frac{\partial k}{\partial \omega} \right|_{\omega_0} x)} \end{aligned} \quad (8.35)$$

We can infer that the wavepacket travels with a velocity determined by

$$\frac{x}{t} = \text{const.} = \left(\left. \frac{\partial k}{\partial \omega} \right|_{\omega_0} \right)^{-1} = \left. \frac{\partial \omega}{\partial k} \right|_{\omega_0} \quad (8.36)$$

This corresponds to the group velocity $v_{gr} = (\frac{\partial \omega}{\partial k})^{-1}$. It varies particularly strong around the resonance $\Delta = 0$. However, at the same time n'' , which describes the absorption, is maximal for $\Delta = 0$. One therefore cannot consider this situation as a real propagation of light since most of it is absorbed. The notion of a group velocity is then not very useful.

- “Spectral hole burning”: $|\Omega_0|^2$ is proportional to the light intensity. Hence, for high intensities absorption (and all other effects) is strongly suppressed even if $\Delta = 0$. The reason is simply that a high intensity corresponds to many photons inside the medium. If all atoms have already been excited by a photon, the remaining photons cannot be absorbed anymore.

8.3 Electromagnetically induced transparency and dark states

The refraction index for atoms with more than two levels can be very different from the two-level result. One example is the phenomenon of electromagnetically induced transparency for a three-level atom in Λ configuration. To derive the index of refraction we again have to solve the Maxwell-Bloch equations with

$$\hat{H} = \hbar \begin{pmatrix} 0 & -\Omega_- & -\Omega_+ \\ -\Omega_-^* & \Delta_- & 0 \\ -\Omega_+^* & 0 & \Delta_+ \end{pmatrix} \quad \mathcal{L}_{\text{sp. Em.}} \hat{\varrho} = \begin{pmatrix} -\gamma \varrho_{ee} & -\frac{\gamma}{2} \varrho_{e-} & -\frac{\gamma}{2} \varrho_{e+} \\ -\frac{\gamma}{2} \varrho_{-e} & \frac{\gamma}{2} \varrho_{ee} & 0 \\ -\frac{\gamma}{2} \varrho_{+e} & 0 & \frac{\gamma}{2} \varrho_{ee} \end{pmatrix} \quad (8.37)$$

Master equation:

$$\dot{\varrho} = -\frac{i}{\hbar} [H, \varrho] + \mathcal{L}_{\text{sp. Em.}} \varrho \stackrel{!}{=} 0 \quad \text{stationary solution} \quad (8.38)$$

To find the solution of this system of linear equations is simple in principle, but to reduce the size of the result we consider the case that Ω_+ is small and expand the solution around $\Omega_+ = 0$.

For $\Omega_+ = 0$, the solution is simply the dark state, i.e.,

$$\varrho_0 = |+\rangle\langle +| \quad (8.39)$$

Since $|+\rangle$ is the dark state for the situation $\Omega_+ = 0$ and $\Omega_- \neq 0$, the entire population is pumped into the state $|+\rangle$.

If we switch on a weak second laser beam ($\Omega_+ \neq 0$) we can calculate its effect using perturbation theory: $\hat{H} = \hat{H}_0 + \hat{H}_1$ with

$$\hat{H}_0 = \hbar \begin{pmatrix} 0 & -\Omega_- & 0 \\ -\Omega_-^* & \Delta_- & 0 \\ 0 & 0 & \Delta_+ \end{pmatrix} \quad \hat{H}_1 = \hbar \begin{pmatrix} 0 & 0 & -\Omega_+ \\ 0 & 0 & 0 \\ -\Omega_+^* & 0 & 0 \end{pmatrix} \quad (8.40)$$

Inserting the ansatz $\hat{\varrho} = \hat{\varrho}_0 + \hat{\varrho}_1 + O(\Omega_+^2)$ into (8.38) we find

$$0 = -\frac{i}{\hbar} [H_0 + H_1, \varrho_0 + \varrho_1] + \mathcal{L}_{\text{sp. Em.}} \hat{\varrho}_0 + \mathcal{L}_{\text{sp. Em.}} \hat{\varrho}_1 \quad (8.41)$$

Since ϱ_0 is the solution of the unperturbed equation, we find for the equation to first order in H_1

$$-\frac{i}{\hbar}[H_0, \varrho_1] + \mathcal{L}_{sp.Em.}\hat{\varrho}_1 = \frac{i}{\hbar}[H_1, \varrho_0] \quad (8.42)$$

This is an inhomogeneous linear system of equations for $\hat{\varrho}_1$. The solution is given by

$$\varrho = \underbrace{|+\rangle\langle+|}_{\varrho_0} + \{\varrho_{e+}|e\rangle\langle+| + \varrho_{-+}|-\rangle\langle+| + \text{H.c.}\} \quad (8.43)$$

with

$$\begin{aligned} \varrho_{e+} &= -\frac{(\Delta_- - \Delta_+)\Omega_+}{(\Delta_- - \Delta_+)(\Delta_+ + i\frac{\gamma}{2}) + |\Omega_-|^2} \\ \varrho_{-+} &= \frac{-\Omega_+\Omega_-^*}{(\Delta_- - \Delta_+)(\Delta_+ + i\frac{\gamma}{2}) + |\Omega_-|^2} \end{aligned}$$

We now calculate the index of refraction for the Ω_+ beam with the aid of Eq. (8.9). We know from the derivation of the two-level refraction index in Sec. 8.2 that³

$$n_+ \approx 1 + \frac{\bar{\varrho}\mathcal{D}}{2\hbar\varepsilon_0} \text{Tr}(\hat{\mathbf{d}}^{(-)} \cdot \boldsymbol{\varepsilon}_{\Omega_+}^* \varrho) \frac{1}{\Omega_+} \quad (8.44)$$

For the Ω_+ beam we have $\hat{\mathbf{d}}^{(-)} \cdot \boldsymbol{\varepsilon}_{\Omega_+}^* \propto |+\rangle\langle e|$, so that

$$n_+ \approx 1 - \frac{\bar{\varrho}\mathcal{D}^2}{2\hbar\varepsilon_0} \frac{\Delta_- - \Delta_+}{(\Delta_- - \Delta_+)(\Delta_+ + i\frac{\gamma}{2}) + |\Omega_-|^2} \quad (8.45)$$

The prefactor $\bar{\varrho}\mathcal{D}^2/(2\hbar\varepsilon_0)$ is typical for optically thin gases and also appears in the two-level result. The term after the prefactor is responsible for EIT.

For $\Delta_- = \Delta_+$ we have $n = 1$, i.e., the same refraction index as in free space; in particular, there is no absorption and the phase velocity is c . However, the system does not completely behave as a vacuum since the right-hand side is trivial only exactly at resonance. The group velocity $\frac{\partial\omega}{\partial k}$ is different from its vacuum value.

Let us consider the case $\Delta_- = 0$ (pump laser in resonance):

$$n_+ = 1 - \frac{\bar{\varrho}\mathcal{D}^2}{2\hbar\varepsilon_0} \frac{(\omega_0 - \omega_+)}{(\omega_0 - \omega_+)(\omega_+ - \omega_0 + i\frac{\gamma}{2}) + |\Omega_-|^2} \quad (8.46)$$

We have for the wave vector

$$k_+(\omega_+) = \frac{\omega_+}{c} n_+(\omega_+) \quad (8.47)$$

³The derivation of this equation is completely analogous to that of Eq. (8.31).

It is then easy to calculate the group velocity,

$$\left. \frac{\partial k_+}{\partial \omega_+} \right|_{\omega_+ = \omega_0} = \frac{n_+}{c} - \frac{\omega_+}{c} \frac{\partial n_+}{\partial \omega_+} = \dots = \frac{1}{c} + \frac{\omega_0}{c} \frac{\bar{\rho} \mathcal{D}^2}{2\hbar \varepsilon_0} \frac{1}{|\Omega_-|^2} \quad (8.48)$$

$$\Rightarrow v_{\text{gr}} = \frac{c}{1 + \frac{\bar{\rho} \mathcal{D}^2}{2\hbar \varepsilon_0} \frac{\omega_0}{|\Omega_-|^2}} \quad (8.49)$$

In principle this would allow us to achieve $v_{\text{gr}} = 0$ (for $\Omega_- \rightarrow 0$). However, in this limit the expansion ($\Omega_+ \ll \Omega_-$) we have made is not valid anymore. Nevertheless, this limit has been achieved experimentally.

Example $\omega_0 = 10^{15} \text{ s}^{-1}$, $\mathcal{D} = ea_0$, $\bar{\rho} \approx 10^{21} \text{ m}^{-3}$, $\Omega_- \approx 10^8 \text{ s}^{-1}$. This results in a group velocity of

$$v_{\text{gr}} = \frac{1}{3} 10^{-6} c \approx 100 \frac{\text{m}}{\text{s}} \quad (8.50)$$

The first experiment by the group of Lene Hau [21] achieved $v_{\text{gr}} = 17 \text{ m/s}$ in 1999. A year later the group of Philipps sogar was able to stop light completely [34]. Stopping of light can be understood as a transfer of atoms from $|+\rangle$ to $|-\rangle$ followed by switching off the pump laser. The information about the probe pulse is then coherently and reversibly stored in the medium. The Ω_+ pulse can be restored after up to a millisecond [35].

Chapter 9

Photonic band gaps

Photonic band gaps (PBG) are completely analogous to the energy bands of electrons in a periodic potential which are well-known in solid state physics. Their physical meaning is that in a periodic dielectric photons cannot propagate at certain frequencies (inside a band gap). One can create such band gaps by a periodic modulation of the index of refraction. To simplify the notation we introduce the relative dielectric constant ε_r ,

$$\varepsilon_r(\mathbf{x}) = \frac{\varepsilon(\mathbf{x})}{\varepsilon_0} = n^2(\mathbf{x}) \quad n: \text{index of refraction} \quad (9.1)$$

which we assume to be time-independent ($\dot{\varepsilon}_r \propto \dot{\varepsilon}(\mathbf{x}) = 0$). We then have

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon(\mathbf{x}) \mathbf{E} = \varepsilon_0 \varepsilon_r(\mathbf{x}) \mathbf{E}(\mathbf{x}) \quad (9.2)$$

and Maxwell's equations become

$$\dot{\mathbf{B}} = -\text{curl } \mathbf{E} \quad \dot{\mathbf{D}} = \varepsilon_0 c^2 \text{curl } \mathbf{B} \quad (9.3)$$

which leads to a “symmetric” wave equation for \mathbf{B} :

$$\left(\partial_t^2 + c^2 \text{curl } \frac{1}{\varepsilon_r(\mathbf{x})} \text{curl} \right) \mathbf{B}(\mathbf{x}, t) = 0 \quad (9.4)$$

Consider now a periodic index of refraction (or dielectric factor) $\varepsilon_r(x) = \varepsilon_r(x + L)$, which for simplicity only depends on x ¹. To emphasize the analogy between photonic and electronic band gaps we will treat the wave equation with concepts borrowed from quantum mechanics.

¹In the following we will nevertheless use the notation $\varepsilon_r(\mathbf{x})$ for a dependence on all components if the equation is generally valid.

- We first define a conserved scalar product (see also Sec. 2.1.6), which will allow us to introduce Hermitean operators:

$$\langle \mathbf{B} | \mathbf{B}' \rangle := -i \frac{\varepsilon_0}{\hbar} \int d^3x (\dot{\mathbf{B}}^* \mathbf{B}' - \mathbf{B}^* \dot{\mathbf{B}}') \quad (9.5)$$

$$\Rightarrow \partial_t \langle \mathbf{B} | \mathbf{B}' \rangle \propto \int d^3x (\ddot{\mathbf{B}}^* \mathbf{B}' - \mathbf{B}^* \ddot{\mathbf{B}}') \quad (9.6)$$

inserting this into the wave equation (9.4)

$$= \int d^3x \left[(-c^2 \text{curl} \frac{1}{\varepsilon_r(\mathbf{x})} \text{curl} \mathbf{B}^*) \mathbf{B}' - \mathbf{B}^* (-c^2 \text{curl} \frac{1}{\varepsilon_r(\mathbf{x})} \text{curl} \mathbf{B}') \right] \quad (9.7)$$

$$= \int d^3x \left[\mathbf{B}^* (-c^2 \text{curl} \frac{1}{\varepsilon_r(\mathbf{x})} \text{curl} \mathbf{B}') - \mathbf{B}^* (-c^2 \text{curl} \frac{1}{\varepsilon_r(\mathbf{x})} \text{curl} \mathbf{B}') \right] = 0 \quad (9.8)$$

In the last step we have performed two partial integrations to move the curl operator to the primed part. We also used that $\varepsilon_r(\mathbf{x})$ does not depend on time and appears between the two curl operators.

- The “Hamiltonian” operator $H := -c^2 \text{curl} \varepsilon_r(\mathbf{x})^{-1} \text{curl}$ is Hermitean, i.e., $\langle \mathbf{B}' | H \mathbf{B} \rangle = \langle H \mathbf{B}' | \mathbf{B} \rangle$. In addition, H is periodic, $H(\mathbf{x} + \mathbf{L}) = H(\mathbf{x})$.

To derive the eigenmodes of the wave equation (9.4) we proceed analogously to solid state theory and first consider the Translation operator translation operator

$$T_{\mathbf{x}_0} = e^{i\hat{\mathbf{p}} \cdot \mathbf{x}_0 / \hbar} = e^{\mathbf{x}_0 \cdot \nabla_{\mathbf{x}}} \quad (9.9)$$

$$\text{Effect of operator: } (T_{\mathbf{x}_0} f)(\mathbf{x}) = f(\mathbf{x} + \mathbf{x}_0) \quad (9.10)$$

$$\begin{aligned} \text{Proof: } T_{\mathbf{x}_0} f(\mathbf{x}) &= e^{\mathbf{x}_0 \cdot \nabla_{\mathbf{x}}} f(\mathbf{x}) \\ &= \sum_{n=0}^{\infty} \frac{(\mathbf{x}_0 \cdot \nabla_{\mathbf{x}})^n}{n!} f(\mathbf{x}) \equiv \text{Taylor-Reihe um } \mathbf{x} \\ &= f(\mathbf{x} + \mathbf{x}_0) \end{aligned} \quad (9.11)$$

H commutes with T_L ,

$$(HT_L - T_L H)f(\mathbf{x}) = Hf(\mathbf{x} + \mathbf{L}) - H(\mathbf{x} + \mathbf{L})f(\mathbf{x} + \mathbf{L}) = 0 ,$$

because H is periodic. Hence, H and T_L share a common system of eigenstates.

The eigenstates of T_L are the momentum states e^{ikx} with eigenvalues $T_L e^{ikx} = e^{ik(x+L)} = e^{ikL} e^{ikx}$. This means the eigenvalues are degenerate for all momenta of the form $k = q + \frac{2n\pi}{L}$, $n \in \mathbb{Z}$ with eigenvalue $\exp(iqL)$. The eigenstates of H can be written as a superposition of degenerate eigenstates of T_L ,

$$\psi_q(x) = e^{iqx} \sum_{n=-\infty}^{\infty} \psi_{q,n} e^{i\frac{2n\pi}{L}x} \quad (9.12)$$

The quantum number q can be chosen to be in the interval $[-\frac{\pi}{2}, \frac{\pi}{2}]$. q is called *quasi momentum* (= momentum modulo $2\pi/L$).

Eigenvalue problem: $\left(\frac{\partial^2}{\partial t^2} - H\right)\psi = 0$, Ansatz $\psi(x, t) = e^{\pm i\omega t}\psi(x) \Rightarrow -\omega^2\psi(x) = H\psi(x)$

Here we consider the most simple case:

$$\epsilon_r = 1 + \delta\epsilon \cos\left(\frac{2\pi}{L}x\right)$$

with $\delta\epsilon \ll 1 \Rightarrow$

$$\begin{aligned} H &= -c^2 \text{curl} \frac{1}{1 + \delta\epsilon \cos\left(\frac{2\pi}{L}x\right)} \text{curl} \\ &\approx -c^2 \text{curl} \left(1 - \delta\epsilon \cos\left(\frac{2\pi}{L}x\right)\right) \text{curl} \\ &= H_0 + \delta\epsilon H_1 \end{aligned}$$

with

$$\begin{aligned} H_0 &= -c^2 \text{curl} \text{curl} \\ H_1 &= c^2 \text{curl} \cos\left(\frac{2\pi}{L}x\right) \text{curl} \end{aligned}$$

Consider a light wave along the x axis,

$$\mathbf{B}(\mathbf{x}) = B(x)\mathbf{e}_y$$

This ansatz fulfills the Maxwell equation $\text{div} \mathbf{B} = 0$. The action of the Hamiltonian becomes

$$\left(-\text{curl} \frac{1}{\epsilon_r(x)} \text{curl} \mathbf{B}\right) = \left(\frac{\partial}{\partial x} \frac{1}{\epsilon_r(x)} \frac{\partial}{\partial x} B\right) \mathbf{e}_y$$

²Another choice can be made by changing the summation index. For instance, for $q' = q + 2\pi/L$ we get the same expansion as above if $\psi_{q,n}$ is replaced by $\psi_{q',n-1}$.

We now employ perturbation theory, $B(x) = \langle x|B\rangle$, $|B\rangle = |B_0\rangle + \delta\epsilon |B_1\rangle$, where $|B_0\rangle$ is a momentum eigenstate p_0 .

$(H_0 + \delta\epsilon H_1) (|B_0\rangle + \delta\epsilon |B_1\rangle) = -\omega^2 (|B_0\rangle + \delta\epsilon |B_1\rangle)$ with $\omega^2 = \omega_0^2 + \delta\epsilon\omega_1^2$
Equation for zeroth order:

$$\begin{aligned} H_0 |p_0\rangle &= -\omega_0^2 |p_0\rangle \\ &= -c^2 \frac{p_0^2}{\hbar^2} |p_0\rangle \end{aligned}$$

$$\Rightarrow \omega_0 = \left| \frac{cp_0}{\hbar} \right|$$

Equation for 1st order:

$$H_0 |B_1\rangle + H_1 |B_0\rangle = -\omega_1^2 |B_0\rangle - \omega_0^2 |B_1\rangle$$

To solve these equations is a bit cumbersome. The structure of the solution is as follows. The change of the eigenvalue can be derived from $-\omega_1^2 = \langle B_0|H_1|B_0\rangle$. The state has the form

$$\begin{aligned} |B_1\rangle &\sim H_1 |B_0\rangle \\ &\sim \text{rot} \cos\left(\frac{2\pi}{L}x\right) \text{rot} |p_0\rangle \\ &\sim \left(e^{i\frac{2\pi}{L}x} + e^{-i\frac{2\pi}{L}x}\right) |p_0\rangle \\ &\sim |p_0 + \underbrace{\frac{2\pi}{L}\hbar}_k\rangle + |p_0 - \frac{2\pi}{L}\hbar\rangle \end{aligned}$$

$$\begin{aligned} H_0 |p\rangle &= -\frac{c^2 p^2}{\hbar^2} |p\rangle \\ &= \omega_0^2(p) |p\rangle \end{aligned}$$

For $p_0 \approx \frac{\hbar k}{2}$ the component $c|p_0 - \hbar k|$ is nearly resonant to $c|p_0|$ while $p_0 + \hbar k$ has a much higher energy $\Rightarrow H_1$ effectively only couples p_0 to $p_0 - \hbar k$ for $p_0 \approx \frac{\hbar k}{2}$ ³. We therefore obtain an effective two-level system. If $|p_0|$ is very different from $\frac{\hbar k}{2}$, coupling is negligible. The energy bands are shown in Fig. 9.2.

Consider the two-level system for $p_0 \approx \frac{\hbar k}{2}$. Denoting the matrix element $\langle p_0|H_1|p_0 - \hbar k\rangle$ as \hbar_1 one obtains the equations

$$|B\rangle \approx \alpha |p_0\rangle + \beta |p_0 - \hbar k\rangle$$

³or p_0 and $p_0 + \hbar k$ for $p_0 \approx -\frac{\hbar k}{2}$

$$\delta\epsilon H_1 |p_0\rangle \approx \delta\epsilon h_1 |p_0 - \hbar k\rangle$$

$$\delta\epsilon H_1 |p_0 - \hbar k\rangle \approx \delta\epsilon h_1 |p_0\rangle$$

and the eigenvalue problem

$$H |B\rangle = -\omega^2 |B\rangle$$

$$H |B\rangle = \begin{pmatrix} -\omega_0^2(p_0) & \delta\epsilon h_1 \\ \delta\epsilon h_1^\dagger & -\omega_0^2(p_0 - \hbar k) \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix}$$

with eigenvalues

$$-\frac{\omega_0^2(p_0) + \omega_0^2(p_0 - \hbar k)}{2} \pm \frac{1}{2} \sqrt{(\omega_0^2(p_0) - \omega_0^2(p_0 - \hbar k))^2 + 4\delta\epsilon^2}$$

where the “detuning” is given by $\omega_0^2(p_0) - \omega_0^2(p_0 - \hbar k) \sim 2\frac{c^2}{\hbar^2}k\Delta p$ in which the momentum $p_0 = \frac{\hbar k}{2} + \Delta p$ is expressed relative to $\hbar k/2$. Then Δp plays a similar role as the real detuning appearing in the two-level system of Sec. 6.3. By varying Δp one obtains, in complete analogy to Landau-Zener transitions, an avoided crossing. This is the reason for the appearance of band gaps.

Significance of photonic band gaps:

- Light with frequency inside a band gap cannot propagate through the medium. This can be used for mirrors, glass fibres, filters, ...
- Theoretical predictions: spontaneous emission of atoms placed in the periodic medium will be strongly modified [22]. However, photonic crystals with band gaps stretching over the entire angular range of photon momenta have only been produced recently [23].

Chapter 10

Appendices

10.1 Green's functions

Consider a general linear inhomogeneous differential equation (DEQ) of the form

$$(\hat{L}_{t,\mathbf{x}}F)(t, \mathbf{x}) = h(t, \mathbf{x}) .$$

It is not important that t and \mathbf{x} represent space and time, they can be replaced by any number of independent variables. A Green's function is a special solution with the inhomogeneity $\delta(\mathbf{x} - \mathbf{x}')\delta(t - t')$.

$$(\hat{L}_{t,\mathbf{x}}G)(t, \mathbf{x}; t', \mathbf{x}') = \delta(\mathbf{x} - \mathbf{x}')\delta(t - t') .$$

The Green's function serves two puposes:

1. If one knows $G(t, \mathbf{x}; t', \mathbf{x}')$, the solution of the general inhomogeneous DEQ is given by

$$F(t, \mathbf{x}) = \int G(t, \mathbf{x}; t', \mathbf{x}') \cdot h(t', \mathbf{x}') d\mathbf{x}' dt' \quad (10.1)$$

Proof:

$$\begin{aligned} (\hat{L}_{t,\mathbf{x}}F)(t, x) &= \hat{L}_{t,\mathbf{x}} \int d\mathbf{x}' dt' G(t, x; t', x') \cdot h(t', x') \\ &= \int d\mathbf{x}' dt' \hat{L}_{t,\mathbf{x}} G(t, x; t', x') \cdot h(t', x') \\ &= \int d\mathbf{x}' dt' \delta(\mathbf{x} - \mathbf{x}')\delta(t - t') h(t', x') = h(t, x) \end{aligned}$$

2. The Green's function can also be used to solve the initial value problem

$$(\hat{L}_{t,\mathbf{x}}F)(t, \mathbf{x}) = 0 \quad , \quad F(t_0, \mathbf{x}) = F_0(\mathbf{x}) . \quad (10.2)$$

Suppose the Green's function fulfills at equal times

$$G(t, x; t, x') = \delta(\mathbf{x} - \mathbf{x}') . \quad (10.3)$$

Then

$$F(t, \mathbf{x}) = \int G(t, \mathbf{x}; t_0, \mathbf{x}') \cdot F_0(\mathbf{x}') d\mathbf{x}' \quad (10.4)$$

Proof:

$$\begin{aligned} (\hat{L}_{t,\mathbf{x}} F)(t, x) &= \int d\mathbf{x}' \hat{L}_{t,\mathbf{x}} G(t, x; t_0, x') \cdot F_0(x') \\ &= \int d\mathbf{x}' \delta(\mathbf{x} - \mathbf{x}') \delta(t - t_0) F_0(x') = 0 \forall t > t_0 \end{aligned}$$

In addition,

$$\begin{aligned} F(t_0, x) &= \int d\mathbf{x}' G(t_0, x; t_0, x') \cdot F_0(x') \\ &= \int d\mathbf{x}' \delta(\mathbf{x} - \mathbf{x}') F_0(x') = F_0(x) . \end{aligned}$$

This concludes the proof.

It is often not too hard to find some Green's function for a given problem. for $t \neq t'$ and $x \neq x'$ it simply fulfills the homogeneous equation. One therefore can often compose G by using different homogeneous solutions for $t < t'$ and $t > t'$. As a rule of thumb, if the highest derivative that appears in \hat{L} is of order n , the $n - 1$ th derivative of G must be discontinuous in order to cancel the delta distribution inhomogeneity.

There are many different Green's functions for a given problem which differ by their boundary conditions. Basically, the choice of homogeneous solutions away from the delta inhomogeneity is fixed by the boundary conditions. One often is especially interested in finding the retarded (or advanced) Green's function. In this case, the $n - 1$ th time derivative is usually proportional to the step function $\theta(t - t')$ (or $\theta(t' - t)$). Bear in mind that the derivative of the step function is the delta distribution.

10.1.1 Green's operators

In a more formal way, Green's functions are actually the integral kernel of a linear operator that acts on a Hilbert space. This is not often appreciated, but it is implicit in its DEQ due to the appearance of the delta distribution. This implies that G itself is also a distribution, and this actually fits nicely to its usual applications.

Considering G as a distribution means that we can only consider integrals like

$$(\hat{G}(t, t')f)(x) := \int dx' G(t, x; t', x') f(t', x') . \quad (10.5)$$

This equation defines a linear operator \hat{G} which maps $f(t', x')$ to $f(t, x)$. This operator has some nice formal features. If Eq. (10.3) is valid then

$$\begin{aligned} (\hat{G}(t, t)f)(x) &= \int dx' G(t, x; t, x') f(t', x') \\ &= \int dx' \delta(x - x') f(t, x') \\ &= \mathbf{1}f(t, x) . \end{aligned} \quad (10.6)$$

Hence, $\hat{G}(t, t) = \mathbf{1}$.

Likewise, one can introduce the operator

$$(\hat{G}h)(t, x) := \int dx' dt' G(t, x; t', x') h(t', x') . \quad (10.7)$$

which leads to

$$\begin{aligned} (\hat{L}_{t, \mathbf{x}} \hat{G}h)(t, x) &= \hat{L}_{t, \mathbf{x}} \int dx' dt' G(t, x; t', x') h(t', x') \\ &= \int dx' dt' \hat{L}_{t, \mathbf{x}} G(t, x; t', x') h(t', x') \\ &= \int dx' dt' \delta(\mathbf{x} - \mathbf{x}') \delta(t - t') h(t', x') \\ &= h(t, x) \end{aligned} \quad (10.8)$$

which means $\hat{L}\hat{G} = \mathbf{1}$. The Green's operator is therefore formally the inverse of the DEQ operator. However, one has to be careful with this interpretation. Every solution of the DEQ corresponds to an eigenfunction of \hat{L} with eigenvalue zero. Hence, this operator is not invertible and the above interpretation is only formal.

An example for Green's operators can be constructed for the Schrödinger equation. The equation

$$(i\partial_t - H)\hat{G}(t, t') = \delta(t - t') \quad (10.9)$$

has the retarded solution

$$\hat{G}(t, t') = -i\theta(t - t')e^{-i(t-t')H} \quad (10.10)$$

Proof:

$$\begin{aligned}
 (i\partial_t - H)(-i)\theta(t-t')e^{-i(t-t')H} &= \delta(t-t')e^{-i(t-t')H} \\
 &\quad - iH\theta(t-t')e^{-i(t-t')H} - H(-i)\theta(t-t')e^{-i(t-t')H} \\
 &= \delta(t-t')e^{-i(t-t')H} \\
 &= \delta(t-t')
 \end{aligned} \tag{10.11}$$

The initial condition is $\hat{G}(t, t) = -i1$ if we set $\theta(0) = 1$. The corresponding Green's function can be derived from

$$\begin{aligned}
 \psi(t, x) &= \langle x | \hat{G}(t, t_0) | \psi(t_0) \rangle \\
 &= \langle x | \int dx' dx'' | x' \rangle \langle x' | \hat{G}(t, t_0) | x'' \rangle \langle x'' | \psi(t_0) \rangle \\
 &= \int dx'' \langle x | \hat{G}(t, t_0) | x'' \rangle \langle x'' | \psi(t_0) \rangle \\
 &= \int dx'' G(t, x; t_0, x'') \psi(t_0, x'')
 \end{aligned} \tag{10.12}$$

where we have set $G(t, x; t_0, x'') = \langle x | \hat{G}(t, t_0) | x'' \rangle$.

10.2 Angular momentum algebra

At the beginning a word of caution: in angular momentum algebra different conventions are used in different textbooks, which are distinguished by their choice of phases of the angular momentum eigenstates. We follow the convention of Ref. [4], which also seems to be used in the Mathematica software package. Other good references on angular momentum in quantum physics are Refs. [2] und [3].

Angular momentum is defined as a vector \mathbf{L} whose three components are operators fulfilling the commutation relations

$$[\mathbf{L}_a, \mathbf{L}_b] = i\hbar \varepsilon_{abc} \mathbf{L}_c. \tag{10.13}$$

It can be shown that $[\mathbf{L}^2, \mathbf{L}_i] = 0$ ist. Hence, one can find a common set of eigenstates of \mathbf{L}^2 and one of the components, which usually is chosen to be L_z :

$$\mathbf{L}^2 |l, m\rangle = \hbar^2 l(l+1) |l, m\rangle, \quad L_z |l, m\rangle = \hbar m |l, m\rangle \tag{10.14}$$

Here, m runs from $-l$ to l and $l = 0, 1/2, 1, \dots$

It is advantageous to introduce the operators $\mathbf{L}_\pm := \mathbf{L}_x \pm i\mathbf{L}_y$, which obey $[\mathbf{L}_\pm, \mathbf{L}_z] = \mp \hbar \mathbf{L}_\pm$ and $[\mathbf{L}_+, \mathbf{L}_-] = 2\hbar \mathbf{L}_z$. It follows that

$$\begin{aligned}
 \mp \hbar \mathbf{L}_\pm |l, m\rangle &= [\mathbf{L}_\pm, \mathbf{L}_z] |l, m\rangle \\
 &= m\hbar \mathbf{L}_\pm |l, m\rangle - \mathbf{L}_z \mathbf{L}_\pm |l, m\rangle
 \end{aligned} \tag{10.15}$$

Hence, $\mathbf{L}_\pm |l, m\rangle$ is proportional to $|l, m \pm 1\rangle$. Since $\mathbf{L}_+^\dagger = \mathbf{L}_-$ and $\mathbf{L}^2 = \mathbf{L}_z^2 + \mathbf{L}_- \mathbf{L}_+ + \hbar \mathbf{L}_z$ we find

$$\begin{aligned} \langle l, m | \mathbf{L}^2 | l, m \rangle &= \hbar^2 l(l+1) \\ &= \langle l, m | \mathbf{L}_z^2 + \mathbf{L}_- \mathbf{L}_+ + \hbar \mathbf{L}_z | l, m \rangle \\ &= \hbar^2 m^2 + \hbar^2 m + |\langle l, m+1 | \mathbf{L}_+ | l, m \rangle|^2 \end{aligned} \quad (10.16)$$

and thus

$$\langle l', m' | \mathbf{L}_+ | l, m \rangle = \langle l, m | \mathbf{L}_- | l', m' \rangle = \hbar f_{l,m} \delta_{l,l'} \delta_{m+1,m'} \quad (10.17)$$

The phase factors of the states $|l, m\rangle$ can be chosen in such a way that the matrix elements are real. We have introduced the notation

$$f_{l,m} := \sqrt{l(l+1) - m(m+1)} \quad (10.18)$$

since this factor appears frequently. A very useful consequence of Eq. (10.17) is that for a given l all the states $|l, m\rangle$ can be obtained from $|l, -l\rangle$ by repeated application of the operator J_+ .

10.2.1 Addition of two angular momenta

When adding two (commuting) angular momentum operators \mathbf{L} and \mathbf{S} to $\mathbf{J} = \mathbf{L} + \mathbf{S}$, one has to address the question of choosing a suitable basis for the product of the respective Hilbert spaces. A possible choice would obviously be the tensor product of the individual bases,

$$|l, m_l; s, m_s\rangle := |l, m_l\rangle \otimes |s, m_s\rangle. \quad (10.19)$$

However, it is usually more convenient to work with eigenstates of \mathbf{J}^2 and \mathbf{J}_z instead. Because $[\mathbf{L}^2, \mathbf{J}_i] = [\mathbf{S}^2, \mathbf{J}_i] = 0$ these states are characterized by four quantum numbers

$$|j, m_j, (l, s)\rangle. \quad (10.20)$$

The matrix elements $\langle l, m_l; s, m_s | j, m_j, (l, s) \rangle =: \langle l, m_l; s, m_s | j, m_j \rangle$ are called *Clebsch-Gordan coefficients* and allow to switch from one basis to the other:

$$|j, m_j, (l, s)\rangle = \sum_{m_l, m_s} \langle l, m_l; s, m_s | j, m_j \rangle |l, m_l; s, m_s\rangle \quad (10.21)$$

By applying \mathbf{J}_z one can easily see that $m_s + m_l = m_j$ is valid. The numerical values for Clebsch-Gordan coefficients can be derived from the linear system of

equations

$$\begin{aligned}
\langle l, m_l; s, m_s | \mathbf{J}_+ | j, m_j, (l, s) \rangle &= \hbar f_{j, m_j} \langle l, m_l; s, m_s | j, m_j + 1 \rangle \quad (10.22) \\
&= \langle l, m_l; s, m_s | (\mathbf{L}_+ + \mathbf{S}_+) | j, m_j, (l, s) \rangle \\
&= \langle j, m_j, (l, s) | (\mathbf{L}_- + \mathbf{S}_-) | l, m_l; s, m_s \rangle^* \\
&= \hbar f_{l, m_l - 1} \langle l, m_l - 1; s, m_s | j, m_j \rangle \\
&\quad + \hbar f_{s, m_s - 1} \langle l, m_l; s, m_s - 1 | j, m_j \rangle
\end{aligned}$$

and the analogous equation for J_- ,

$$\begin{aligned}
f_{j, m_j - 1} \langle l, m_l; s, m_s | j, m_j - 1, \rangle &= f_{l, m_l} \langle l, m_l + 1; s, m_s | j, m_j \rangle \quad (10.23) \\
&\quad + f_{s, m_s} \langle l, m_l; s, m_s + 1 | j, m_j \rangle,
\end{aligned}$$

as well as the normalization condition

$$\begin{aligned}
1 &= \langle j, m_j, (l, s) | j, m_j, (l, s) \rangle \\
&= \sum_{m_l, m_s} |\langle l, m_l; s, m_s | j, m_j \rangle|^2. \quad (10.24)
\end{aligned}$$

Generally, Clebsch-Gordan coefficients are only non-zero for $l + s \geq j \geq |l - s|$. In the literature one also finds Wigner's $3j$ -symbol instead of Clebsch-Gordan coefficients, to which they are related by

$$\begin{pmatrix} l & s & j \\ m_l & m_s & -m_j \end{pmatrix} := \frac{(-1)^{l-s+m_j}}{\sqrt{2j+1}} \langle l, m_l; s, m_s | j, m_j \rangle. \quad (10.25)$$

Wigner's $3j$ -symbol is more symmetric than Clebsch-Gordan coefficients. It is invariant under cyclic permutations of the three columns. Under an anti-cyclic permutation, or under simultaneous change of the sign of m_l , m_s and m_j , it obtains a factor of $(-1)^{l+s+j}$.

10.2.2 Tensor operators and Wigner-Eckart theorem

Tensor operators T_{k, m_k} are defined by their transformation properties under rotations. They correspond to an irreducible representation of the rotation group. Equivalently, they can be defined as operators which fulfill the following commutation relations with a given angular momentum operator \mathbf{J} ,

$$[J_z, T_{k, m_k}] = \hbar m_k T_{k, m_k} \quad (10.26)$$

$$[J_+, T_{k, m_k}] = \hbar f_{k, m_k} T_{k, m_k + 1} \quad (10.27)$$

$$[J_-, T_{k, m_k}] = \hbar f_{k, m_k - 1} T_{k, m_k - 1}. \quad (10.28)$$

Tensor operators are of great importance for atomic selection rules and allow an elegant derivation of the latter by means of the Wigner-Eckart theorem.

To prove the Wigner-Eckart theorem we first consider the states

$$|j'', \widetilde{m_j''}(j, k)\rangle := \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle T_{k, m_k} |j, m_j\rangle \quad (10.29)$$

The action of J_z on these states is given by

$$\begin{aligned} J_z |j'', \widetilde{m_j''}(j, k)\rangle &= \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle \times \\ &\quad ([J_z, T_{k, m_k}] + T_{k, m_k} J_z) |j, m_j\rangle \\ &= \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle \times \\ &\quad (\hbar m_k T_{k, m_k} + T_{k, m_k} \hbar m_j) |j, m_j\rangle \\ &= \hbar m_j'' |j'', \widetilde{m_j''}(j, k)\rangle \end{aligned} \quad (10.30)$$

where the last step follows from the fact that Clebsch-Gordan coefficients are non-zero only for $m_k + m_j = m_j''$. The action of J_+ reads

$$\begin{aligned} J_+ |j'', \widetilde{m_j''}(j, k)\rangle &= \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle ([J_+, T_{k, m_k}] + T_{k, m_k} J_+) |j, m_j\rangle \\ &= \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle \times \\ &\quad \left(\hbar f_{k, m_k} \underbrace{T_{k, m_k + 1}}_{=m_k'} |j, m_j\rangle + T_{k, m_k} \hbar f_{j, m_j} \underbrace{|j, m_j + 1\rangle}_{=m_j'} \right) \\ &= \hbar \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k - 1 \rangle f_{k, m_k - 1} T_{k, m_k} |j, m_j\rangle \\ &\quad + \hbar \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j - 1; k, m_k \rangle f_{j, m_j - 1} T_{k, m_k} |j, m_j\rangle \\ &= \hbar \sum_{m_k, m_j} (\langle j'', m_j'' | j, m_j; k, m_k - 1 \rangle f_{k, m_k - 1} \\ &\quad + \langle j'', m_j'' | j, m_j - 1; k, m_k \rangle f_{j, m_j - 1}) T_{k, m_k} |j, m_j\rangle \\ &= \hbar \sum_{m_k, m_j} f_{j'', m_j''} \langle j'', m_j'' + 1 | j, m_j; k, m_k \rangle T_{k, m_k} |j, m_j\rangle \\ &= \hbar f_{j'', m_j''} |j'', \widetilde{m_j'' + 1}(j, k)\rangle \end{aligned} \quad (10.31)$$

where we have used Eq. (10.23) in the second-to-last step. Analogously, we find for J_-

$$J_- |j'', \widetilde{m_j''}(j, k)\rangle = \hbar f_{j'', m_j''-1} |j'', \widetilde{m_j''-1}(j, k)\rangle \quad (10.32)$$

The action of the angular momentum operators on the states $|j'', \widetilde{m_j''}(j, k)\rangle$ is the same as on the angular momentum eigenstates $|j'', m_j''\rangle$. Therefore, these states need to be proportional to each other. Since for given j'' the eigenstates $|j'', m_j''\rangle$ can be constructed by applying J_+ to $|j'', -j''\rangle$, this can also be done for the states $|j'', \widetilde{m_j''}(j, k)\rangle$. For this reason, the proportionality factor between the states is independent from m_j'' and we find

$$|j'', \widetilde{m_j''}(j, k)\rangle = (-1)^{2k} \frac{\langle j'' || T_k || j \rangle}{\sqrt{2j''+1}} |j'', m_j''\rangle. \quad (10.33)$$

The symbol $\langle j'' || T_k || j \rangle$ is called reduced matrix element and contains the dependence of the proportionality factor on j'', k and j . The factor of $(-1)^{2k}/\sqrt{2j''+1}$ is convention. It is easy to see that the reduced matrix element can be calculated by

$$\langle j'' || T_k || j \rangle = (-1)^{2k} \sqrt{2j''+1} \sum_{m_k, m_j} \langle j'', m_j'' | j, m_j; k, m_k \rangle \langle j'', m_j'' | T_{k, m_k} | j, m_j \rangle \quad (10.34)$$

The matrix elements of tensor operators can be derived from Eq. (10.33). Because of the completeness of the angular momentum eigenstates we have

$$\begin{aligned} \sum_{j'', m_j''} \langle j, m_j; k, m_k | j'', m_j'' \rangle |j'', \widetilde{m_j''}(j, k)\rangle &= \sum_{j'', m_j''} \langle j, m_j; k, m_k | j'', m_j'' \rangle \times \\ &\quad \sum_{m_k', m_j'} \langle j'', m_j'' | j, m_j'; k, m_k' \rangle T_{k, m_k'} |j, m_j'\rangle \\ &= \sum_{m_k', m_j'} \langle j, m_j; k, m_k | j, m_j'; k, m_k' \rangle T_{k, m_k'} |j, m_j'\rangle \\ &= T_{k, m_k} |j, m_j\rangle \end{aligned} \quad (10.35)$$

and thus

$$\begin{aligned} \langle j', m_j' | T_{k, m_k} | j, m_j \rangle &= \sum_{j'', m_j''} \langle j, m_j; k, m_k | j'', m_j'' \rangle \langle j', m_j' | j'', \widetilde{m_j''}(j, k) \rangle \\ &= (-1)^{2k} \frac{\langle j' || T_k || j \rangle}{\sqrt{2j'+1}} \langle j, m_j; k, m_k | j', m_j' \rangle \end{aligned} \quad (10.36)$$

This is the Wigner-Eckart theorem.

10.2.3 Addition of three angular momenta

When adding three or more angular momenta one has to be careful with the choice of basis. Consider the case $\mathbf{F} = \mathbf{I} + \mathbf{L} + \mathbf{S}$. If we first add $\mathbf{J} = \mathbf{S} + \mathbf{L}$ and then $\mathbf{F} = \mathbf{I} + \mathbf{J}$, the procedure of the previous section would result in the basis states $\{|f, m_f, (i, j(s, l))\rangle\}$, i.e., eigenstates of \mathbf{F}^2 , \mathbf{F}_z , \mathbf{I}^2 , and \mathbf{J}^2 . However, if we first add $\mathbf{\Sigma} = \mathbf{I} + \mathbf{S}$ and then $\mathbf{F} = \mathbf{\Sigma} + \mathbf{L}$, we implicitly would use the basis $\{|f, m_f, (\sigma(i, s), l)\rangle\}$. The two sets of basis vectors are different. In a sense the addition of angular momenta is therefore not associative (although the addition of the operators is of course associative). Which of the two bases is more suitable depends entirely on the physical situation at hand.

Detailed construction of the basis states:

$$\begin{aligned}
 |f, m_f, (i, j(s, l))\rangle &= \sum_{m_i, m_j} \langle i, m_i; j, m_j | f, m_f \rangle |i, m_i\rangle \otimes |j, m_j(s, l)\rangle \\
 &= \sum_{m_i, m_j} \langle i, m_i; j, m_j | f, m_f \rangle \times \\
 &\quad \sum_{m_s, m_l} \langle s, m_s; l, m_l | j, m_j \rangle |i, m_i\rangle \otimes |s, m_s\rangle \otimes |l, m_l\rangle
 \end{aligned}
 \tag{10.37}$$

$$\begin{aligned}
 |f, m_f, (\sigma(i, s), l)\rangle &= \sum_{m_\sigma, m_l} \langle \sigma, m_\sigma; l, m_l | f, m_f \rangle |\sigma, m_\sigma(i, s)\rangle \otimes |l, m_l\rangle \\
 &= \sum_{m_\sigma, m_l} \langle \sigma, m_\sigma; l, m_l | f, m_f \rangle \times \\
 &\quad \sum_{m_i, m_s} \langle i, m_i; s, m_s | \sigma, m_\sigma \rangle |i, m_i\rangle \otimes |s, m_s\rangle \otimes |l, m_l\rangle
 \end{aligned}
 \tag{10.38}$$

Multiplication of the vectors results in

$$\begin{aligned}
\langle f, m_f, (i, j(s, l)) | f', m'_f, (\sigma(i, s), l) \rangle &= \sum_{\substack{m_j, m_\sigma \\ m_i, m_l, m_s}} \langle i, m_i; j, m_j | f, m_f \rangle \langle \sigma, m_\sigma; l, m_l | f', m'_f \rangle \\
&\quad \langle s, m_s; l, m_l | j, m_j \rangle \langle i, m_i; s, m_s | \sigma, m_\sigma \rangle \\
&= (2f+1) \sqrt{(2j+1)(2\sigma+1)} \sum_{\substack{m_j, m_\sigma \\ m_i, m_l, m_s}} (-1)^{2i-j-2l+\sigma+2m} \\
&\quad \begin{pmatrix} i & j & f \\ m_i & m_j & -m_f \end{pmatrix} \begin{pmatrix} \sigma & l & f' \\ m_\sigma & m_l & -m'_f \end{pmatrix} \\
&\quad \begin{pmatrix} s & l & j \\ m_s & m_l & -m_j \end{pmatrix} \begin{pmatrix} i & s & \sigma \\ m_i & m_s & -m_\sigma \end{pmatrix} \\
&=: \delta_{f,f'} \delta_{m_f, m'_f} \sqrt{(2j+1)(2\sigma+1)} \times \\
&\quad (-1)^{i+s+l+f} \left\{ \begin{matrix} i & s & \sigma \\ l & f & j \end{matrix} \right\}
\end{aligned}$$

The last line defines Wigner's $6j$ -Symbol, which agrees with the so-called Racah symbols $W(\dots)$ up to a sign:

$$\left\{ \begin{matrix} i & s & \sigma \\ l & f & j \end{matrix} \right\} = (-1)^{i+s+l+f} W(i, s, l, f; \sigma, j). \quad (10.40)$$

The expression for the $6j$ -Symbol as a sum over Clebsch-Gordan coefficients is equivalent to Eq. (C.32) of Ref. [4].

10.2.4 Matrix elements for composed systems

The content of this section is of great importance for the actual calculation of multipole matrix elements for atoms and molecules. One often has to resolve the problem that the atomic energy eigenstates are (to a good approximation) eigenstates of the hyperfine-spin operator F , but the dipole matrix elements have to be calculated with respect to the electronic states alone (without nuclear spin I). Generally one needs to express the reduced matrix element $\langle f'(i', j') || T_k^{(j)} || f(i, j) \rangle$ of a tensor operator $T_{k, m_k}^{(j)}$, that only acts on the total electronic angular momentum J , through $\langle j' || T_k^{(j)} || j \rangle$.

To achieve this one starts from Eq. (10.34) and expands, with the aid of Eq. (10.21), the hyperfine states $|f, m_f\rangle$ in terms of the nuclear spin eigenstates $|i, m_i\rangle$ and the eigenstates $|j, m_j\rangle$ of the total electronic angular momentum. One

then arrives at

$$\begin{aligned}
\langle f'(i', j') || T_k^{(j)} || f(i, j) \rangle &= (-1)^{2k} \sqrt{2f' + 1} \sum_{m_k, m_f} \langle f', m'_f | f, m_f; k, m_k \rangle \langle f', m'_f | T_{k, m_k}^{(j)} | f, m_f \rangle \\
&= (-1)^{2k} \sqrt{2f' + 1} \sum_{m_k, m_f} \sum_{m_i, m_j} \sum_{m'_i, m'_j} \langle f', m'_f | f, m_f; k, m_k \rangle \langle f', m'_f | i', m'_i; j', m'_j \rangle \\
&\quad \langle i, m_i; j, m_j | f, m_f \rangle \langle i', m'_i, j', m'_j | T_{k, m_k}^{(j)} | i, m_i, j, m_j \rangle \\
&= (-1)^{2k} \sqrt{2f' + 1} \sum_{m_k, m_f} \sum_{m_i, m_j} \sum_{m'_i, m'_j} \langle f', m'_f | f, m_f; k, m_k \rangle \langle f', m'_f | i', m'_i; j', m'_j \rangle \\
&\quad \delta_{i, i'} \delta_{m_i, m'_i} \langle i, m_i; j, m_j | f, m_f \rangle \langle j', m'_j | T_{k, m_k}^{(j)} | j, m_j \rangle \\
&= \sqrt{2f' + 1} \delta_{i, i'} \sum_{\substack{m_k, m_f \\ m_i, m_j, m'_j}} \langle f', m'_f | f, m_f; k, m_k \rangle \langle f', m'_f | i', m'_i; j', m'_j \rangle \times \\
&\quad \langle i, m_i; j, m_j | f, m_f \rangle \frac{\langle j' || T_k^{(j)} || j \rangle}{\sqrt{2j' + 1}} \langle j, m_j; k, m_k | j', m'_j \rangle \\
&= \sqrt{\frac{2f' + 1}{2j' + 1}} \delta_{i, i'} \langle j' || T_k^{(j)} || j \rangle \times \\
&\quad \sum_{\substack{m_k, m_f \\ m_i, m_j, m'_j}} \langle f, m_f; k, m_k | f', m'_f \rangle \langle i, m_i; j', m'_j | f', m'_f \rangle \times \\
&\quad \langle i, m_i; j, m_j | f, m_f \rangle \langle j, m_j; k, m_k | j', m'_j \rangle
\end{aligned} \tag{10.4}$$

The sum in the last two lines can be written as a $6j$ symbol by using Eq. (10.39) with $f' = f, m'_f = m_f$. In the sum over the Clebsch-Gordan coefficients only m_f is not a summation index. Therefore f in Eq. (10.39) corresponds to f' in Eq. (10.41). All other relations can be derived from the position of the indices in the Clebsch-Gordan coefficients. One thus deduces

$$\begin{aligned}
\left\{ \begin{array}{ccc} i & j & f \\ k & f' & j' \end{array} \right\} &= \frac{(-1)^{i+j+k+f'}}{\sqrt{(2j' + 1)(2f + 1)}} \sum_{\substack{m'_j, m_f \\ m_i, m_k, m_j}} \langle i, m_i; j', m'_j | f', m'_f \rangle \langle f, m_f; k, m_k | f', m'_f \rangle \\
&\quad \langle j, m_j; k, m_k | j', m'_j \rangle \langle i, m_i; j, m_j | f, m_f \rangle
\end{aligned} \tag{10.42}$$

and therefore

$$\begin{aligned}
\langle f'(i', j') || T_k^{(j)} || f(i, j) \rangle &= \sqrt{(2f + 1)(2f' + 1)} \delta_{i, i'} (-1)^{i+j+k+f'} \times \\
&\quad \langle j' || T_k^{(j)} || j \rangle \left\{ \begin{array}{ccc} i & j & f \\ k & f' & j' \end{array} \right\}
\end{aligned} \tag{10.43}$$

This is equivalent to Eq. (C.90) of Ref. [4].

10.3 Distributions

Distributions are mathematical objects which, strictly speaking, are only defined if they appear in an integral. For instance, $\delta(x - x_0)$ is only defined through its effect inside the integral

$$\int dx f(x) \delta(x - x_0) := f(x_0)$$

Here, $f(x)$ is a so-called Test function. These are functions which, speaking in a sloppy way, “always behave well”. More formally, they are C^∞ functions with compact support, or at least they converge sufficiently fast to zero for $|x| \rightarrow \infty$). Distributions are no functions. For example, $\delta(x - x_0)$ is not well defined as a function at the position x_0 .

The integral $D(\omega) := \int_0^\infty d\tau e^{-i\omega\tau}$ is also only defined in the sense of distributions:

$$\begin{aligned} \int_{-\infty}^{\infty} d\omega D(\omega) f(\omega) &= \int_{-\infty}^{\infty} d\omega f(\omega) \int_0^\infty d\tau e^{-i\omega\tau} \\ &= \int_{-\infty}^{\infty} d\omega f(\omega) (\pi\delta(\omega) - i\frac{P}{\omega}) \\ &= \pi f(0) - i \lim_{\epsilon \rightarrow 0^+} \left(\int_{-\infty}^{-\epsilon} \frac{d\omega}{\omega} f(\omega) + \int_{\epsilon}^{\infty} \frac{d\omega}{\omega} f(\omega) \right) \end{aligned}$$

The transition from the first to the second line relies on a mathematical theorem. The principal value in this expression means that one has to use the same small parameter ϵ on both sides of the singularity of the integrand. If one calculates the limiting value of an integral without principal value, one has to admit different small parameters on both sides of the singularity.

Bibliography

- [1] J.D. Jackson, *Classical Electrodynamics*, Wiley 1999.
- [2] J.J. Sakurai, *Modern Quantum Mechanics*, Addison Wesley, 1994.
- [3] D.M. Brink and G.R. Satchler, *Angular Momentum*, Oxford Library of Physical Sciences 1968.
- [4] A. Messiah, *Quantum mechanics*, Vol. 2, Wiley, New York 1963.
- [5] R. F. Streater and A. S. Wightman, *PCT, spin and statistics, and all that* (1964) Beautiful text on the axiomatic formulation of QFT, i.e., the derivation of the properties of any QFT from a few basic axioms (such as Einstein locality). Mathematically superb, although some conclusions of this theory are discouraging. It nevertheless provides rigorous proofs of the spin-statistics theorem (i.e, that Bosons must have integer spin) and the PCT theorem (any QFT must be invariant under a space inversion followed by charge conjugation and time reversal).
- [6] D.F. Walls and G. Milburn, *Quantum Optics*, Springer, Berlin (1995).
- [7] P. Meystre and M. Sargent, *Elements of Quantum Optics*, Springer, Berlin (1999).
- [8] P.W. Milonni, *The quantum vacuum : an introduction to quantum electrodynamics* (1994). A very interesting book which mostly deals with the effect of boundaries (Casimir effect) and some particular quantum field theoretical effects in (mostly non-relativistic) QED.
- [9] J.D.Bjorken and S.D.Drell *Relativistic Quantum Fields*, Mcgraw-Hill (1965).
- [10] E.R. Pike and S. Sarkar, *The quantum theory of radiation*, Clarendon Press, Oxford (1995).

- [11] C. Cohen-Tannoudji, J. Dupont-Roc and G. Grynberg, *Atom-photon interactions : basic processes and applications* (1992). A very good book dealing with the fundamental techniques to describe the interaction of atoms with light. Includes dressed states, resolvent method, master equations.
- [12] C. Cohen-Tannoudji, *Atomic motion in laser light*, in *Fundamental systems in quantum optics*, edited by J. Dalibard, J.M. Raimond and J. Zinn-Justin, Les Houches Session LIII, Elsevier Science Publishers 1992.
- [13] J. Dalibard and C. Cohen-Tannoudji, *Atomic motion in laser light: connection between semiclassical and quantum descriptions*, J. Phys. B **18**, 1661 (1985).
- [14] S.-Y. Zhu and M.O. Scully, *Spectral Line Elimination and Spontaneous Emission Cancellation via Quantum Interference*, Phys. Rev. Lett. **76**, 388 (1996).
- [15] H.-R. Xia, C.-Y. Ye, and S.-Y. Zhu, *Experimental observation of spontaneous emission cancellation*, Phys. Rev. Lett. **77**, 1032 (1996).
- [16] Li Li, X. Wang, J. Yang, G. Lazarov, J. Qi, and A. M. Lyyra, *Comment on "Experimental Observation of Spontaneous Emission Cancellation"*, Phys. Rev. Lett. **84**, 4016 (2000).
- [17] M. Abramowitz and I.A. Stegun (Eds.), *Handbook of mathematical functions*, Dover, New York 1972.
- [18] A. Galindo and P. Pascual, *Quantum mechanics*, Springer, Heidelberg (1990).
- [19] M. Redhead, *Incompleteness, nonlocality and realism : a prolegomenon to the philosophy of quantum mechanics*, Oxford: Clarendon Pr., 1987.
- [20] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, *Laser cooling below the one photon recoil energy by velocity-selective coherent population trapping: theoretical analysis*, Journ. Opt. Soc. Am. B **6**, 2112 (1989).
- [21] L.V. Hau., SE Harris, Z. Dutton, and C.H. Behroozi, *Light speed reduction to 17 metres per second in an ultracold atomic gas*, Nature **397**, 594-598 (1999).
- [22] S. John and T. Quang, *Spontaneous emission near the edge of a photonic band gap*, Phys. Rev. A **50**, 1764 (1994).

- [23] A. Blanco *et al.*, *Nature* **405**, 437 (2000).
- [24] S. Grossmann, *Funktionalanalysis*, Frankfurt a.M.: Akad.Verl.ges., 1970.
- [25] Y. Castin, *Bose-Einstein condensates in atomic gases: simple theoretical results*, <http://xxx.uni-augsburg.de/cond-mat/0105058>.
- [26] M. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, *Science* **269**, 198 (1995).
- [27] M.-O. Mewes, M. R. Andrews, N. J. van Druten, D. M. Kurn, D. S. Durfee, C. G. Townsend and W. Ketterle, *Phys. Rev. Lett.* **77**, 416 (1996).
- [28] E.A. Power and S. Zienau, *Coulomb gauge in nonrelativistic quantum electrodynamics and the shape of spectral lines*, *Phil. Trans. A* **251**, 427 (1959).
- [29] R.G. Woolley, *A reformulation of molecular quantum electrodynamics*, *Journ. Phys. B* **7**, 488 (1974).
- [30] Claude Cohen-Tannoudji, Jacques Dupont-Roc, and Gilbert Grynberg, *Photons and atoms : introduction to quantum electrodynamics (1989)*. In this book the fundamental equations of non-relativistic QED are thoroughly explained.
- [31] D.P. Craig and T. Thirunamachandran, *Molecular quantum electrodynamics : an introduction to radiation-molecule interactions (1997)*. Despite its title it is also very useful for atoms. A cheap (15 Euro) alternative to the first book.
- [32] M.O. Scully and M.S. Zubairy, *Quantum Optics (1997)*. A very nice introduction which also covers electromagnetically induced transparency.
- [33] Leonard Mandel and Emil Wolf, *Optical coherence and quantum optics (1995)*. One of the most comprehensive and complete texts on quantum optics. Gives a good survey of most techniques and topics.
- [34] D.F. Phillips, A. Fleischhauer, A. Mair, R. L. Walsworth, M. D. Lukin, *Storage of Light in Atomic Vapor*, *Phys. Rev. Lett.* **86**, 783 (2001).
- [35] C. Liu, Z. Dutton, C.H. Behrooz, and L.V. Hau, *Observation of coherent optical information storage in an atomic medium using halted light pulses*, *Nature* **409**, 490 (2001).
- [36] F. Riehle, T. Kisters, A. Witte, J. Helmcke and C.J. Bordé, *Optical Ramsey spectroscopy in a rotating frame: Sagnac effect in a matter-wave interferometer*, *Phys. Rev. Lett.* **67**, 177 (1991).

- [37] C.J. Bordé, *Atomic interferometry with internal state labeling*, Phys. Lett. A **140**, 10 (1989).
- [38] M. Kasevich and S. Chu, *Atomic Interferometry using stimulated Raman Transitions*, Phys. Rev. Lett. **67**, 181 (1991).
- [39] O. Carnal and J. Mlynek, *Young's Double Slit Experiment with Atoms: A Simple Atom Interferometer*, Phys. Rev. Lett. **66**, 2689 (1991).
- [40] T. Kato, J. Phys. Soc. Jap. **5**, 435 (1950).
- [41] M.V. Berry, Proc. Roy. Soc. (Lond.) **392**, 45 (1984).
- [42] Y. Aharonov and D. Bohm, *Significance of electromagnetic potentials in quantum theory*, Phys. Rev., **115**, 485-491 (1959).
- [43] A. Galindo and P. Pascual, *Quantum mechanics II*, Springer, Berlin 1991.
- [44] Lewis H. Ryder, *Quantum field theory* (1985).
- [45] Y. Aharonov and D. Rohrlich, *Quantum Paradoxes: Quantum Theory for the Perplexed*, Wiley-VCH, 2004.
- [46] M. F. Andersen, C. Ryu, Pierre Clade, V. Natarajan, A. Vaziri, K. Helmerson, and W. D. Phillips, *Quantized Rotation of Atoms from Photons with Orbital Angular Momentum*, Phys. Rev. Lett. **97**, 170406 (2006).
- [47] K.-P. Marzlin, W. Zhang, and E.M. Wright, *Vortex Coupler for Atomic Bose-Einstein Condensates*, Phys. Rev. Lett. **79**, 4728 (1997).
- [48] C. K. Hong, Z. Y. Ou, and L. Mandel, *Measurement of subpicosecond time intervals between two photons by interference*, Phys. Rev. Lett. **59**, 2044 (1987).
- [49] I. L. Chuang and Y. Yamamoto, *Simple quantum computer*, Phys. Rev. A **52**, 3489 (1995).
- [50] Z.-B. Wang, K.-P. Marzlin and B. C. Sanders, *Large Cross-Phase Modulation between Slow Copropagating Weak Pulses in ^{87}Rb* , Phys. Rev. Lett. **97**, 063901 (2006).
- [51] L. Boivin, F. X. Kärtner, and H. A. Haus, *Analytical solution to the quantum field theory of self-phase modulation with a finite response time*, Phys. Rev. Lett. **73**, 240 (1994).
- [52] J. H. Shapiro, *Single-photon Kerr nonlinearities do not help quantum computation*, Phys. Rev. A **73**, 062305 (2006).

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